Estimated Radiological Inventory Sent from Argonne National Laboratory-West to the Subsurface Disposal Area from 1952 through 1993

Michael L. Carboneau James A. Vail

ldaho Completion Proiect

Bechtel BWXT Idaho, LLC

April 2004

Estimated Radiological Inventory Sent from Argonne National Laboratory-West to the Subsurface Disposal Area from 1952 through 1993

Michael L. Carboneau James A. Vail

April 2004

Idaho Completion Project Idaho Falls, Idaho 83415

Prepared for the
U.S. Department of Energy
Assistant Secretary for Environmental Management
Under DOE/NE Idaho Operations Office
Contract DE-AC07-99ID13727

ABSTRACT

This report represents a revised estimate of the inventory of radiological contaminants shipped from 1952 through 1993 from the Argonne National Laboratory-West (ANL-W) facilities and buried in the Subsurface Disposal Area, part of the Radioactive Waste Management Complex at the Idaho National Engineering and Environmental Laboratory. This inventory has been updated to support development of the comprehensive remedial investigation and feasibility study for Operable Unit 7-13/14 under the Comprehensive Environmental Response, Compensation, and Liability Act.

This report describes the methodology for identifying, collecting, compiling, reviewing, and revising the radiological waste inventory from ANL-W. In addition, descriptions of the following are provided: (1) the ANL-W facilities that shipped the waste (i.e., the waste generators), (2) processes by which waste was generated, (3) availability of waste disposal information, (4) sources of data, and (5) approaches for collecting and analyzing these data. The uncertainties associated with these data are mentioned.

ACKNOWLEDGMENTS

The authors are very appreciative of the many people that have contributed to the radioactive inventory analysis and the writing, editing, and review of this report. Without these contributions, this report would not have been possible. First, the authors would like to thank Karen Taylor for her assistance in locating the waste shipment data concerning the ANL-W facilities. Second, we would like to thank Craig Kullberg for his initial analysis and the original draft of this report. Many sections of this final report are based on Craig's work. Third, we would like to thank Roy Grant for his technical review, comments, and recommendations concerning the preliminary and final drafts of this report. Fourth, we would like to thank Ed May for his editorial assistance. Finally, the authors would like to thank Nancy Hampton, K. Jean Holdren, Bruce Becker, and Brandt Meagher for their management oversight of this program.



CONTENTS

ABS	TRAC	Т	iii
ACK	NOW	LEDGMENTS	v
ACR	ONYN	MS	xi
1.	INTI	RODUCTION	1
	1.1	Purpose	1
	1.2	Scope	5
	1.3	Document Organization	5
2.	BAC	KGROUND	7
	2.1	Brief History and Description of the Subsurface Disposal Area	7
	2.2	History and Description of Argonne National Laboratory-West Site and Associated Waste Generators	7
		2.2.1 Experimental Breeder Reactor-II. 2.2.2 Transient Reactor Test Facility. 2.2.3 Zero Power Physics Reactor. 2.2.4 Fuel Conditioning Facility. 2.2.5 Hot Fuel Examination Facility. 2.2.6 Fuel Manufacturing Facility. 2.2.7 Laboratory and Office Building. 2.2.8 Support Facilities. 2.2.9 Other Argonne-Affiliated Test Facilities.	9 9 10 10 10
3.	EST	MATED RADIOLOGICAL INVENTORIES FOR 1952 THROUGH 1983	12
	3.1	Background	12
	3.2	Disposal Waste Streams	14
	3.3	Data Collection and Analysis	17
	3.4	Overview of the Analysis Methodology	17
	3.5	Activation Product Inventory	18
	3.6	Estimated Actinide Inventory	20
	3.7	Estimated Fission Product Inventory	25
	3.8	Uncertainties	28

4.	EST	MATED RADIOLOGICAL INVENTORY FOR 1984 THROUGH 1993	31
	4.1	Background	31
	4.2	Disposal Waste Streams	32
	4.3	Data Collection and Analysis	32
	4.4	Overview of the Analysis Methodology	34
	4.5	Activation Product Inventory	35
	4.6	Estimated Actinide Inventory	36
	4.7	Estimated Fission Product Inventory	39
	4.8	Uncertainties	40
5.	CON	ICLUSIONS AND RECOMMENDATIONS	41
6.	REF.	ERENCES	45
Appe		A—Major Waste Streams Containing Fuel and Miscellaneous Actinides as Reported Historical Data Task and the Recent and Projected Data Task	. A-1
Appe	endix E	3—Details of the Radioactive Source Term Sent from ANL-W to the SDA	. B-1
Appe		C—Reassessment of the Activation Product Inventory Sent to the SDA in EBR-II ssembly Hardware.	C-1
		FIGURES	
1.		of the Idaho National Engineering and Environmental Laboratory showing the location e Radioactive Waste Management Complex and other major facilities	2
2.		of the Radioactive Waste Management Complex showing the location of the Subsurface osal Area	3
3.	Facil	ities at Argonne National Laboratory-West	4
4.	Map	of Argonne National Laboratory-West	4
		TABLES	
1.		s activity for annual disposals in the SDA of the RWMC as reported for the years 1952 ugh 1983 from ANL-W facilities	13
2.		inal and current descriptions for ANL-W waste streams buried in the SDA during the s 1952 through 1983	16
3.	Sum	mary of the analysis methods used to analyze each ANL-W waste stream for the HDT	18

4.	Comparison of activation-product activities for the years 1952 through 1983	. 19
5.	Best-estimate mass inventory of bulk-actinide wastes (heavy metals) with little or no irradiation that were sent to the SDA from 1960 through 1983, and are included in waste-stream ANL-MOD-4H	22
6.	Shipping data for irradiated fuel-bearing wastes (e.g., fuel samples) sent to the SDA from 1960 through 1983 (heavy metals mass data included in ANL-MOD-3H and ANL-MOD-2H)	. 23
7.	Best-estimate actinide disposals made at the SDA from 1960 through 1983	. 24
8.	Summary of the total fission product waste activities that were disposed at the SDA from ANL-W operations from 1960 through 1983	. 27
9.	Irradiated blanket and fuel material from EBR-I for which disposition histories are incomplete or unknown	. 29
10.	Summary of gross activity by year of waste shipped from ANL-W and disposed of at the SDA from 1984 through 1993	31
11.	Original and modified descriptions for ANL-W waste streams buried in the SDA from 1984 to 1993	. 33
12.	Summary of the analysis methods used to analyze each ANL-W waste stream for the RPDT	. 34
13.	Comparison of activation-product activities for the years 1984 through 1993	. 35
14.	Overall comparison of activation-product activities for the years 1952 through 1993	. 36
15.	Best-estimate mass inventory of unirradiated uranium wastes sent to the SDA from 1984 through 1993 (e.g., during the RPDT) and are included in the ANL-MOD-2R waste-stream	. 37
16.	Shipping data for irradiated fuel-bearing wastes (e.g., fuel samples) sent to the SDA from 1984 through 1993	. 37
17.	Best-estimate actinide disposals made at the SDA from 1984 through 1993	. 38
18.	Total actinide disposals made at the SDA from 1960 through 1993	. 38
19.	Summary of the total fission product waste activities that were disposed at the SDA from ANL-W operations for 1984 through 1993 (obtained from B-31)	. 40
20.	Summary of the total inventory of activation products, fission products, and actinide waste activities that were disposed at the SDA from ANL-W for 1960 through 1993	. 42
21.	Best-estimate activity distribution of radionuclides within the ten possible waste streams based on the total curie inventory (1960 through 1993)	. 43
22.	Best-estimate percent distribution of curie inventory within the ten possible waste streams (1960 through 1993)	44

ACRONYMS

AFSR Argonne Fast Source Reactor

ANL-E Argonne National Laboratory-East

ANL-W Argonne National Laboratory-West

BORAX Boiling Water Reactor Test Experiment

COC contaminant of concern

DD&D deactivation, decontamination, and decommissioning

DOE U.S. Department of Energy

EBR-I Experimental Breeder Reactor-I

EBR-II Experimental Breeder Reactor-II

FCF Fuel Conditioning Facility or Fuel Cycle Facility

FMF Fuel Manufacturing Facility

HDT Historical Data Task

HFEF/N Hot Fuel Examination Facility – North

HFEF/S Hot Fuel Examination Facility – South

HM heavy metal

INEEL Idaho National Engineering and Environmental Laboratory

INTEC Idaho Nuclear Technology and Engineering Center

L&O Laboratory and Office Building

LLW low-level waste

MFP mixed fission products

MW Megawatt

MWD Megawatt Day

MW_e Megawatt electric

MW_tD Megawatt thermal Day

RLWTF Radioactive Liquid Waste Treatment Facility

RPDT Recent and Projected Data Task

RWMC Radioactive Waste Management Complex

RWMIS Radioactive Waste Management Information System

SA Subassembly

SCMS Sodium Components Maintenance Shop

SDA Subsurface Disposal Area

SHADE shielded hot-air drum evaporators

TREAT Transient Reactor Test Facility

TRU transuranic

ZPPR Zero Power Physics Reactor

ZPR3 Zero Power Reactor Unit 3

Estimated Radiological Inventory Sent from Argonne National Laboratory-West to the Subsurface Disposal Area from 1952 through 1993

1. INTRODUCTION

This report reflects updates and revisions to the previously documented inventory of radiological contaminants generated at the Argonne National Laboratory-West (ANL-W) facilities and subsequently disposed of in the Subsurface Disposal Area (SDA), a part of the Radioactive Waste Management Complex (RWMC) located at the Idaho National Engineering and Environmental Laboratory (INEEL). The locations of the RWMC, the SDA, ANL-W, and other INEEL facilities are shown in Figure 1. An enlarged view of the SDA is shown in Figure 2. Figure 3 shows the facilities at ANL-W. The objective of this report is to provide a more detailed and complete evaluation of the source term or inventory of radioactive wastes^a that were sent to the SDA from ANL-W facilities since the original estimates were compiled and documented. These estimates were compiled in two periods: (1) 1952 through 1983, known as the "Historical Data Task" (HDT), and (2) 1984 through 1993, known as the "Recent and Projected Data Task" (RPDT) (LMITCO 1995a, 1995b). The former was governed by a variety of waste acceptance criteria, and the latter is covered by the current low-level waste (LLW) performance assessment (Case et al. 2000). Waste was thus disposed of in the SDA under varying requirements for documentation and content.

1.1 Purpose

The objective of compiling these waste inventories is to support development of the comprehensive remedial investigation and feasibility study for the RWMC.^b Locations of RWMC and the SDA are shown on Figures 1 and 2, respectively. Models that will be used to support the risk analysis of the cleanup are based on historical records of inventories and on investigations such as this report.^c Waste streams containing Operable Unit 7-13/14 contaminants of concern (Holdren and Broomfield 2003) and buried in the SDA are being quantified to support investigation required under the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) (42 USC § 9601 et seq. 1980).

a. Inventory estimates for nonradioactive contaminants are not modified.

b. The RWMC is designated Waste Area Group 7 and the comprehensive RI/FS is identified as OU 7-13/14. The RI/FS focuses primarily on the SDA.

c. This study has been developed within the framework of the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) as implemented in the Federal Facility Agreement and Consent Order (FFA/CO) between the U.S. Department of Energy, the Idaho Department of Environmental Quality, and the U.S. Environmental Protection Agency.

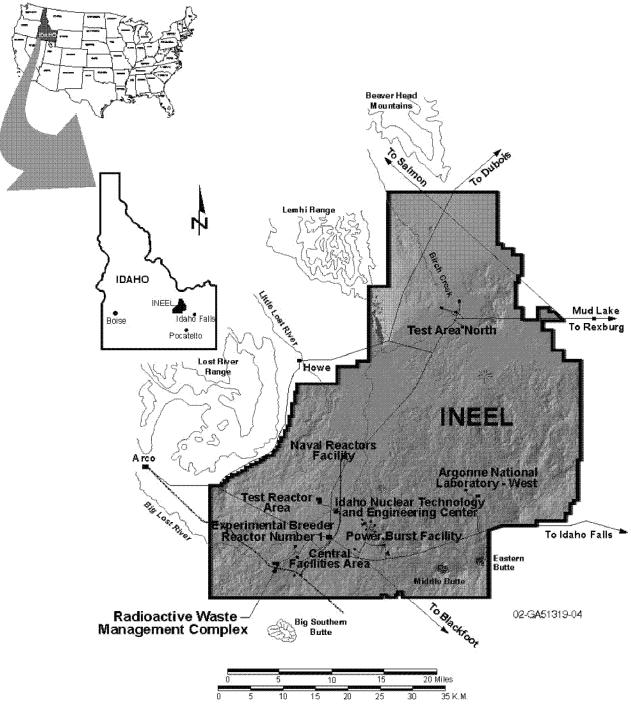


Figure 1. Map of the Idaho National Engineering and Environmental Laboratory showing the location of the Radioactive Waste Management Complex and other major facilities.

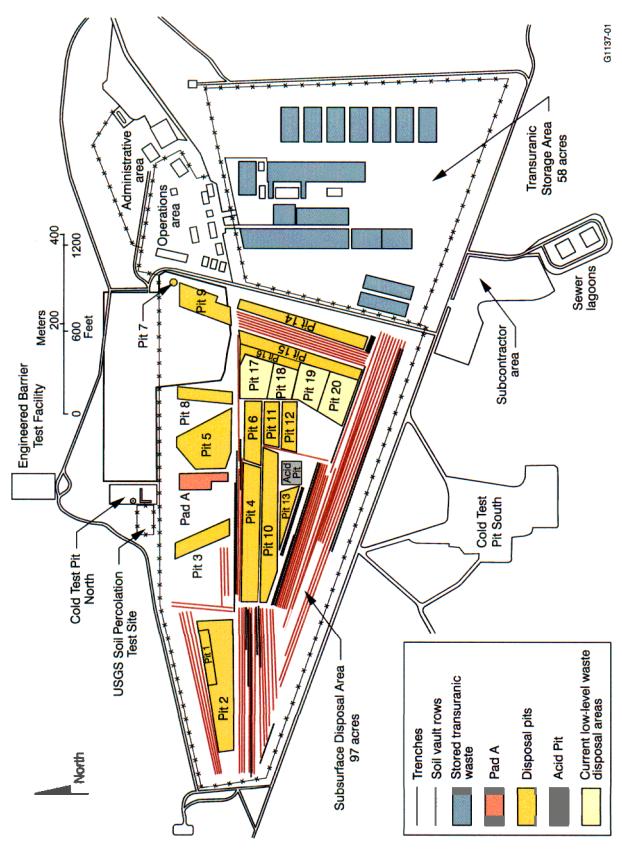


Figure 2. Map of the Radioactive Waste Management Complex showing the location of the Subsurface Disposal Area.



Figure 3. Facilities at Argonne National Laboratory-West.

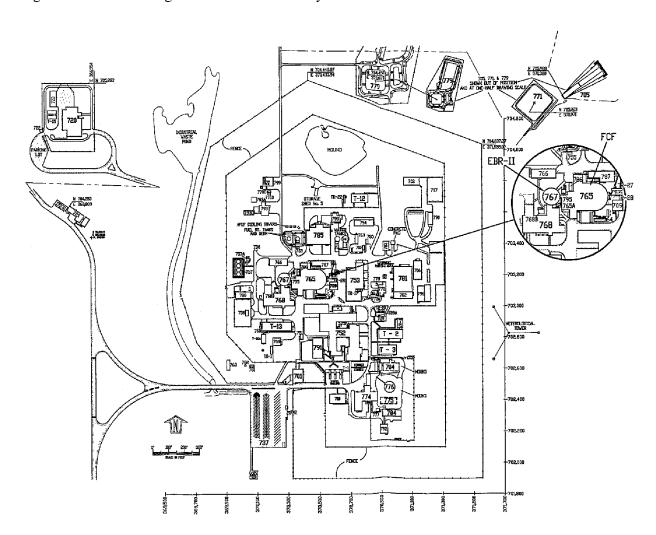


Figure 4. Map of Argonne National Laboratory-West.

1.2 Scope

This report reassesses the disposal histories of radiological contaminants produced at ANL-W facilities and then disposed of at the SDA during the two referenced periods (1952 through 1983 and 1984 through 1993). The original HDT and RPDT did not include estimates for the complete nuclide-specific breakdown for contaminants of concern (COCs) identified in the *Ancillary Basis for Risk Analysis of the Subsurface Disposal Area* (Holdren et al. 2002). The principal contaminants fall into three categories: activation products, fission products, and actinides (i.e., isotopes of uranium and transuranic [TRU] elements). The radiological inventory in this assessment supercedes the radioactive inventory values described in the previous HDT or RPDT reports (LMITCO 1995a, 1995b). However, this report does not consider, nor does it update, the nonradiological inventory compiled for the HDT and RPDT.

Since waste acceptance criteria and record-keeping protocols for the SDA have changed over time, the receipt of radioactive materials buried in the SDA is divided into two periods reflecting these two different disposal histories: (1) 1952 through 1984 and (2) 1985 through 1993. During the latter period, waste disposal in the SDA was limited to low-level radioactive waste from INEEL waste generators; information about handling various types of waste is readily available. Disposal practices have not changed significantly since 1984 and are much more stringent. However, during the former period, routine shallow landfill disposal of radioactive and hazardous waste was the commonly accepted technology of choice. Requirements for documentation were predicated on waste-handling issues rather than on long-term environmental impacts. In particular, at the SDA, TRU and mixed waste—mostly from the Rocky Flats Plant in Colorado—were disposed of through 1970. Low-level mixed waste containing hazardous chemical and radioactive contaminants was normally accepted through 1983. However, these types of wastes are no longer being accepted for routine disposal at the RWMC.

Disposals made after 1993 were reviewed by Little et al. (2001) and are not addressed in this reevaluation. Details of the historical operations at the RWMC, including waste disposal practices, have been previously documented in Section 3 of the *Ancillary Basis for Risk Analysis of the Subsurface Disposal Area* (Holdren et al. 2002) and in the report *A History of the Radioactive Waste Management Complex at the Idaho National Engineering Laboratory* (EG&G 1985).

1.3 Document Organization

The following paragraphs briefly describe the remaining sections in this report:

- Section 2 provides a brief history of the SDA and a description of the ANL-W facilities that are considered under the present reassessment analysis, and some reactors operated by Argonne Laboratory-East (ANL-E) but located on the INEEL site outside the present ANL-W boundaries.
- Section 3 provides the general methods and results of updated radionuclide waste inventory information for ANL-W and some affiliated facilities that shipped waste to the SDA for disposal during the years 1952 through 1983.
- Section 4 provides the general methods and results of updated radionuclide waste inventory information for ANL-W that was shipped to the SDA for disposal during the RPDT period (1984 through 1993).
- Section 5 summarizes the main results of the inventory assessment analysis
- Section 6 lists the references cited throughout this report

- Appendix A provides detailed data concerning heavy-metal disposals made at the SDA from 1960 through 1988. These disposals included irradiated fuel samples (or similar materials) disposed at SDA.
- Appendix B provides the detailed information that defines the radioactive source term sent to the SDA from ANL-W facilities.
- Appendix C provides the detailed activation product inventory data for the subassembly hardware that was irradiated in the EBR-II core and eventually disposed at the SDA (1977 through 1993).

2. BACKGROUND

The following brief history and description of the SDA and ANL-W's waste generating facilities helps make clear the difficulties that were encountered in assessing the source-term inventories, the analysis methodology, and the corresponding activity uncertainties.

2.1 Brief History and Description of the Subsurface Disposal Area

The SDA is a radioactive waste landfill located at the RWMC at the INEEL in southeastern Idaho (Figure 1). Contaminants in the landfill include hazardous chemicals, remote-handled fission and activation products, and TRU radionuclides. Located in the southwestern quadrant of the INEEL, the facility's 72 ha (177 acres) is divided into three separate areas by function: the SDA, the Transuranic Storage Area (TSA), and the administration and operations area.

The original landfill, established in 1952, was originally called the NRTS Burial Ground. Now part of the SDA, the original landfill covered 5.2 ha (13 acres) and was used for shallow-land disposal of solid radioactive waste. In 1958, the disposal area was expanded to 35.6 ha (88 acres). Relocating the security fence in 1988 outside the dike surrounding the disposal area established the SDA's current size as 39 ha (97 acres). The TSA was added to the RWMC in 1970. Located adjacent to the east side of the SDA, the TSA's 23 ha (58 acres) is used to store, prepare, and ship retrievable TRU waste to the Waste Isolation Pilot Plant southeast of Carlsbad, New Mexico. The 9-ha (22-acre) administration and operations area at the RWMC includes administrative offices, maintenance buildings, equipment storage, and miscellaneous support facilities. A map of RWMC disposal locations and facilities is shown in Figure 2.

2.2 History and Description of Argonne National Laboratory-West Site and Associated Waste Generators

Located in the southeastern part of the INEEL, ANL-W is approximately 56 km (35 miles) west of Idaho Falls, Idaho. The ANL-W site was established in 1958 with the construction of the Transient Reactor Test Facility (TREAT). ANL-W is the primary center in the United States for testing and demonstrating nuclear energy technology and experiments. The mission at ANL-W emphasizes technologies associated with nuclear fuel, including advanced methods for fuel reprocessing, improving fuel efficiency, and testing fuel performance. In addition, the ANL-W mission includes technologies for characterizing nuclear material and restoring the environment, and technologies and processes requiring remote handling of nuclear fuel.

The seven major facilities that comprise the ANL-W site and their operational status are listed as follows:

- Experimental Breeder Reactor-II (EBR-II) (1961 1994)
- Transient Reactor Test Facility (TREAT) (1959 presently in standby)
- Zero Power Physics Reactor (ZPPR) (1969 presently in standby). Originally named Zero Power Plutonium Reactor.
- Hot Fuel Examination Facility; formerly called HFEF/N (1972 present)
- Fuel Conditioning Facility (FCF) (1964 present). Originally named Hot Fuel Examination Facility South (HFEF/S).

d. TREAT was constructed at Site 16 of the NRTS (now the INEEL) and is a part of the ANL-W site. Construction was started in February 1958 and completed in November 1958. The reactor achieved criticality on February 23, 1959.

- Fuel Manufacturing Facility (FMF) (1986 currently operating)
- Laboratory and Office (L&O) Building (1962 currently operating).

Other ANL-W support facilities include:

- Radioactive Liquid Waste Treatment Facility (RLWTF) (1983 present)
- Sodium Components Maintenance Shop (SCMS) (1960 operating)
- Sodium Process Facility (1987 in standby)

Additional test reactors (many these have been deactivated, decontaminated and/or decommissioned) that were managed by ANL-E in Illinois^e but were located at the INEEL in regions distant from the ANL-W site, are listed as follows:

- EBR-I (1951 1964)
- Zero Power Reactor Unit 3 (ZPR3) (1955 1970)
- Argonne Fast Source Reactor (AFSR), (1959^f presently defueled)
- Boiling Water Reactor Test Experiments (BORAX-I through BORAX-V) (1953 1964).

2.2.1 Experimental Breeder Reactor-II

The original EBR-II was designed to demonstrate a complete operational breeder reactor power plant with onsite reprocessing of metallic fuel; demonstrations were successfully carried out from 1964 to 1969. EBR-II achieved initial criticality on September 30, 1961 (i.e., without sodium), and "wet" criticality (i.e., with the core submerged in liquid sodium coolant) on November 11, 1963. EBR-II went to power (12 MW_e) on August 13, 1964. The EBR-II facility has: (1) a sodium-cooled reactor with a maximum thermal power rating of 62.5 MW, (2) an



intermediate closed loop of secondary sodium, and (3) a steam plant that produced 20 MW (gross) or 16.5 MW (net) electrical power through a conventional turbine generator.

From 1969, the emphasis at EBR-II shifted to an irradiation facility that tested fuels and materials for future, larger, liquid-metal cooled reactors. The EBR-II facility also provided electrical power for ANL-W and INEEL sites. EBR-II was officially shut down on September 30, 1994. Since then, EBR-II has been prepared for deactivation, decontamination, and decommissioning (DD&D). During its lifetime (August 1, 1964 until September 30, 1994), EBR-II generated 366,780 MWD of thermal energy.

e. Some reactors located at the INEEL site were operated by ANL-E located in Illinois. The ANL-E facilities located on the INEEL site are distinguished from the Argonne reactors located on the present day ANL-W site. This distinction was made to agree with general format of the HDT and RPDT documents. Nevertheless, some confusion and overlap of waste items can occur. For example, waste generated at an ANL-E facility may have been shipped to ANL-W facilities for examination, repackaged, and then disposed of at the SDA.

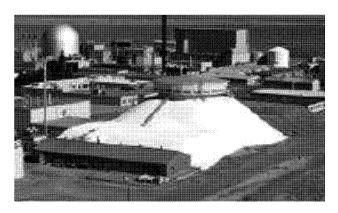
f. The Argonne Fast Source Reactor (AFSR) was moved from its original location near EBR-I to the ANL-W site in 1970.

2.2.2 Transient Reactor Test Facility

Construction of TREAT was started in February 1958 and completed in November 1958. The reactor achieved criticality on February 23, 1959. This reactor was designed to produce short, controlled bursts of nuclear energy to simulate accident conditions leading to nuclear fuel damage. TREAT is an uranium oxide-fueled, graphite-moderated, air-cooled reactor that provided data on fuel cladding damage, fuel motion, coolant channel blockages, molten fuel and coolant interactions, and potential explosive forces during an accident. In 1968, TREAT was used to make high-resolution neutron-radiographs of irradiated fuel and other irradiated specimens. Currently, TREAT is in standby status.

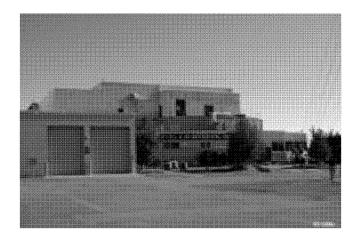
2.2.3 Zero Power Physics Reactor

The ZPPR (formerly the Zero Power Plutonium Reactor) is the national facility for testing the physics properties of advanced, fast-spectrum reactors; it allows study of the properties of experimental reactor cores. Experimental cores are built by hand-loading plates of reactor materials into drawers that are then put into a designed pattern. Reactor core designs are tested at essentially zero power levels. ZPPR is similar in concept to the ZPR-3 reactor. Currently, ZPPR is in standby status.



2.2.4 Fuel Conditioning Facility

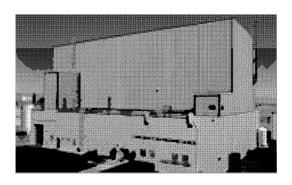
This facility first became operational in 1964 as the Fuel Cycle Facility (FCF) renamed Hot Fuel Examination Facility South (HFEF/S) in 1972 when HFEF-N was built.. HFEF/S has demonstrated pyrometallurgical reprocessing of EBR-II fuel during its first few years of operation. A remotely operated production line was used to reprocess spent EBR-II fuel and return it to the reactor. That mission was discontinued after successful demonstration of the process. In 1996, HFEF/S was renamed FCF.



The FCF has been used to examine irradiated fuels and material experiments from EBR-II and TREAT, and to provide other reactor support services (e.g., transfer of spent fuel to the Idaho Nuclear Technology and Engineering Center [INTEC]). The FCF consists of two hot cells, one with an air atmosphere and the other with an inert argon gas atmosphere. A total of 23 hot cell workstations surround the outer perimeter of the FCF hot cells, and four active workstations make up the workspace of the argon cell. The FCF is currently operational.

2.2.5 Hot Fuel Examination Facility

The HFEF was built in 1972 and became operational in 1975 and is currently used to examine irradiation experiments. Examinations conducted in the HFEF provide essential data for determining the performance and conditions of fuels and materials irradiated in the EBR-II reactor, the TREAT reactor, and other DOE reactor facilities. The HFEF consists of two shielded hot cells:



(1) the decontamination cell, which contains an air atmosphere, and (2) the main cell, which contains an argon gas atmosphere. Each HFEF hot cell is equipped with shielded windows and master-slave manipulators. The main cell is used for work exposing materials that would react chemically with air (e.g., sodium and plutonium).

Neutron Radiography Facility: The Neutron Radiography Facility, a 250-kW training, research, and isotope reactor, is in the HFEF and provides a neutron source for radiography. This facility is equipped with two beam tubes and two separate radiography stations for neutron radiography of irradiated components. Facilities to decontaminate and repair hot cell equipment and manipulators are also in the HFEF. The capability to examine and characterize contact-handled TRU waste (destined for the Waste Isolation Pilot Plant in New Mexico) was added to HFEF in 1990.

2.2.6 Fuel Manufacturing Facility

The FMF contains the entire operation for manufacturing metallic fuel elements within a single building. The building contains a casting furnace and large gloveboxes to encapsulate and bond the cast-fuel slugs in a stainless steel jacket.



2.2.7 Laboratory and Office Building

The analytical laboratory in the L&O Building provides chemistry support for ANL-W in the areas of environmental compliance, fuel chemistry, sodium and water chemistry, and waste classification analysis. The laboratory consists of hot cells and chemistry laboratories.

2.2.8 Support Facilities

- **2.2.8.1** Radioactive Liquid Waste Treatment Facility. In 1983, the RLWTF began receiving liquid LLW from ANL-W facilities to store in tanks before evaporation in the shielded hot-air drum evaporators (SHADE). Before 1983, the low-level liquid evaporation process took place in the basement of the L&O Building. Currently, liquid waste is piped from the L&O Building, the FCF, and the HFEF directly to the RLWTF facility.
- **2.2.8.2 Sodium Components Maintenance Shop.** The SCMS, also associated with the EBR-II support facility, removes sodium from reactor components for repair or replacement.
- **2.2.8.3 Sodium Process Facility.** The function of the SPF is to convert primary and secondary sodium coolant from EBR-II to a chemically stable compound suitable for landfill disposal.

2.2.9 Other Argonne-Affiliated Test Facilities

The ANL-W also served as a trans-shipment site or waste disposal reporting location for waste generated from other Argonne affiliated test facilities located on the INEEL but not at the ANL-W site. These waste streams included material from the liquid metal-cooled EBR-I and the water-cooled BORAX test reactors.

- **2.2.9.1 Experimental Breeder Reactor-I.** The EBR-I was the first reactor built at INEEL. It achieved criticality in 1951 and was the first reactor to produce electric power beginning December 20, 1951. EBR-I operated at a thermal power level of 1.4 MW and produced 250 kW of electricity. This reactor was decommissioned in 1964. EBR-I was a liquid metal-cooled reactor that provided a test bed for numerous fast reactor experiments. These experiments employed highly enriched uranium fuels in the first three test cores and plutonium in the fourth test core. From 1951 until 1962, EBR-I produced ~300 MW_tD of thermal energy. On November 27, 1962, EBR-I achieved criticality with a plutonium core. In conjunction with EBR-I, two other reactor test facilities were housed in the same test complex.
- **2.2.9.2 Zero Power Reactor Unit 3.** The first test facility, the ZPR-3, was a low-power criticality test reactor. This reactor was shut down in 1970.
- **2.2.9.3 Argonne Fast Source Reactor.** The second facility (the AFSR) was a low-power, neutron physics test reactor powered by a highly enriched uranium 21-kg core that employed a depleted neutron reflector. The AFSR was moved from its original location near EBR-I to the ANL-W site in 1970. Currently, the fuel has been removed from the AFSR.
- **2.2.9.4 Boiling Water Reactor Test Experiment.** The BORAX test reactors consisted of five experimental boiling water reactor test facilities. The BORAX-I test reactor began operating in 1953 and the final test reactor, BORAX-V, began operating in 1963. All of these reactors were either decommissioned or shut down after 1964. The BORAX program provided valuable data for proof-of-principle demonstrations that light-water reactors could be built and safely operated.

3. ESTIMATED RADIOLOGICAL INVENTORIES FOR 1952 THROUGH 1983

Waste inventory disposals for the ANL-W facilities were originally compiled for the years 1952 through 1983 in the HDT report (LMITCO 1995a). To be consistent with the previous work, the reassessed inventory was separated into the same general period; however, there are some exceptions. For example, since most of the facilities that are located on the ANL-W site were not in operation until after 1958, it is believed that very little radioactive waste from ANL-W was generated and disposed of during the 1952 through 1959 period. Practically all of the waste sent to the SDA from ANL-W was generated and shipped after 1960. Therefore, most of the early data is limited to the period of 1960 through 1983. Furthermore, except for uranium blanket material from EBR-I, which was disposed at the RWMC in 1964, 1965, and in 1967; and for a small amount of waste from the BORAX facilities, no other radioactive waste from Argonne reactors (or facilities) located outside the present ANL-W site is considered in this report. In other words, the present analysis is mainly concerned with waste shipments made to the SDA from facilities located on the ANL-W site.

3.1 Background

Generally, for the years 1952 through 1983, there was less detailed and less accurate information concerning waste disposal activities than exists in the comparable period of 1984 through 1993 (see Section 4). Therefore, there was less information during the first period to characterize the radioactive waste sent to the SDA. In addition, ANL-W disposals sent to the SDA during the 1952–1983 period had a more complex history in relation to later disposals. The patterns of waste disposal show significant variations as illustrated in Table 1, which presents the gross activity for yearly disposals of radionuclides from ANL-W facilities. Note that disposal records for ANL-W generally do not exist before 1960.^h

During the 1960s, an EBR-II experimental program was started to demonstrate the technical feasibility of reprocessing fast reactor fuels (Stevenson 1987; Koch, Loewenstein, and Monson 1962). From 1964 through 1970, the experimental fuel reprocessing campaign was conducted. Table 1 shows an increase in the gross annual activity at the beginning of this reprocessing campaign and then a decrease at the end of this campaign. In particular, irradiated EBR-II fuel was reprocessed at the ANL-W FCF for recasting into new fuel elements. More than 400 subassemblies containing irradiated and recycled fuel were fabricated for return to the reactor. The three major waste streams associated with this experimental program included significant amounts of fission product-bearing hot cell waste, reprocessing hardware waste, and samples from driver-fuel assemblies. Typically, hot cell waste consisted of fission product-bearing material generated during the reprocessing of irradiated EBR-II driver fuel. Melted-down driver and blanket fuels were cast into metal ingots and then recast into fuel pins.

Waste that was reprocessed consisted of various hardware components used with the casting, melting, and refining equipment to manufacture new fuel, including such items as melt-refining induction furnaces. In addition, a marked increase occurred in disposals of dissolved driver-fuel waste—generally Analytical Laboratory samples stabilized in vermiculite before disposal—during the experimental reprocessing program. Recording contaminant-specific isotopic breakdowns of the above-mentioned waste streams on the SDA shipping manifests was considered impractical because hundreds of these shipments were made. Gross inventory activities from 1960 through 1970 are based on Form 110 shipping manifests.

g It is mentioned in Volume 2 of the HDT report that "... records available at ANL-W show no disposal at RWMC until 1958."

h. Waste Area Group 7 does not currently have pre-1960 shipping manifests. However, this may not be an important factor for ANL-W since most ANL-W facilities were built after 1960.

i. In some cases, ingots of reprocessed driver fuel were sent to INTEC for reprocessing because of contamination by iron impurities.

Table 1. Gross activity for annual disposals in the SDA of the RWMC as reported for the years 1952 through 1983 from ANL-W facilities.

SDA	Gross Annual	
Disposal	Activity	
Year(s)	(Ci)	Important Events
1952–1958		EBR-I is operational.
1959		TREAT (located on the ANL-W site) becomes operational.
1960	1.5E+00	Construction of EBR-II (started in 1957) is continuing.
1961	8.0E+01	EBR-II achieves criticality without sodium.
1962	5.0E+02	_
1963	3.0E+00	EBR-II achieves "wet" criticality with sodium.
1964	4.2E+02	EBR-I is decommissioned. FCF now called HFEF/S becomes
		operational. EBR-II power is raised to 45 MW (thermal) and produces
		$700 \text{ MW}_{t}D.$
1965	3.9E+02	EBR-II produces 4,340 MW _t D.
1966	4.6E+03	EBR-II produces 7,200 MW _t D.
1967	1.1E+04	EBR-II produces 3,300 MW _t D.
1968	2.1E+04	EBR-II produces 7,000 MW _t D.
1969	9.0E+03	EBR-II fuel reprocessing program ended; ZPPR begins operation;
		EBR-II produces 7,900 MW _t D.
1970	2.1E+03	EBR-II reconfigured to be test irradiation facility. New constraints on
		TRU waste disposals begin. EBR-II produces 11,400 MW _t D.
1971	3.6E+03	EBR-II produces 8,700 MW _t D.
1972	4.2E+01	EBR-II produces 10,700 MW _t D.
1973	8.5E+02	EBR-II produces 11,600 MW _t D.
1974	7.4E+02	EBR-II produces 13,300 MW _t D.
1975	1.5E+02	HFEF/N begins operation. EBR-II produces 14,760 MW _t D.
1976	4.3E+02	EBR-II produces 18,100 MW _t D.
1977	1.9E+05	Disposals of irradiated EBR-II subassembly hardware begin in the
		SDA. EBR-II produces 16,400 MW _t D.
1978	1.8E+05	EBR-II produces 16,500 MW _t D.
1979	2.3E+05	EBR-II produces 16,000 MW _t D.
1980	1.3E+05	EBR-II produces 16,300 MW _t D
1981	1.1E+05	EBR-II produces 13,700 MW _t D.
1982	1.7E+05	EBR-II produces 12,100 MW _t D.
1983	4.8E+04	EBR-II produces 12,600 MW _t D.
Total	1.1E+06	-

EBR = Experimental Breeder Reactor

HFEF = Hot Fuel Examination Facility

 MW_tD = Megawatt thermal days

SDA = Subsurface Disposal Area

TREAT = Transient Reactor Test Facility

TRU = transuranic

ZPPR = Zero Power Physics Reactor

After 1969, EBR-II operations were redirected to a variety of experimental irradiation test programs. The EBR-II continued to be the main source of SDA radionuclide contaminants for the remainder of the HDT period. Beginning in 1977, disposals of irradiated subassembly hardware in soil vaults greatly increased the net amount of radionuclide activities going to the SDA. Activation products (e.g., Co-60) were responsible for this sharp increase in reported activities.

Contact-handled LLW was generated by routine facility operations and included metallic, combustible, and noncombustible waste, which originated from the FMF, L&O Building, FCF, HFEF, TREAT, ZPPR, EBR-II, SCMS, and RLWTF facilities. Other facility operations (such as ZPPR) resulted in disposing of bulk amounts of contact-handled unirradiated uranium waste. The relative amount of contact-handled uranium is significantly larger than the limited amounts of uranium-bearing remote-handled waste.

Remote-handled waste originated mainly from FCF and HFEF and consisted of high-gamma-field waste from EBR-II. These gamma fields resulted from Co-60 and other activation materials produced by prolonged neutron irradiation of subassembly hardware in the EBR-II reactor. The irradiated subassembly hardware was separated from irradiated fuel elements in hot cells at the HFEF and eventually disposed of in SDA soil vaults. A much smaller fraction of the remote-handled waste consisted of fission products and TRU nuclides. In some cases, remote-handled waste records showed that irradiated fuel samples were either shipped separately or with activated subassembly hardware. Moreover, some of the subassembly hardware had traces of TRU and fission-product contamination. This contamination was the consequence of hot cell disassembly activities or other operations.

However, amounts of TRU contaminants and fission products buried in the SDA dramatically decreased after 1970. This reduction coincides with the termination of the EBR-II fuel reprocessing campaign and the introduction of more stringent restrictions on the types of waste sent to the SDA. Net activities for these waste categories from 1971 through 1983 are based on numerical totals calculated from the Radioactive Waste Management Information System (RWMIS) database. One major source of radionuclides came from the destructive examination of a variety of experimental fuel elements from EBR-II. The radionuclides of interest were fission products and TRU contaminants associated with these examinations.

Between 1977 and 1983, disposal activities at the SDA were similar to post-1983 patterns. That is, high-activity waste streams coming from examinations of irradiated fuel and irradiated subassembly hardware were managed as remote-handled waste and buried in the SDA soil vaults. In 1977, disposals began of activated EBR-II subassembly hardware. Shipping records and information from ANL-W personnel indicate that no irradiated EBR-II assembly hardware was sent to the SDA before 1977.

3.2 Disposal Waste Streams

The original waste stream definitions from the HDT report (LMITCO 1995a), and the modified definitions for the years 1952–1983 are shown in Table 2. The principal contaminants fall into three categories: activation products, fission products, and actinides (including TRU isotopes). Although the waste streams originally identified for this period were reasonably complete, high- and low-activity waste were generally combined (see ANL-765-2H), and thus the distinction was not clear between high-activity and low-activity waste in many of those records. Second, dominant contaminant groupings (such as TRU content) were not always clearly identified.

The high-activity disposals of irradiated subassemblies during the years 1977–1983 previously identified as contained in waste streams ANL-765-2H and ANL-785-1H were combined into a single

j. Available records of remote-handled waste show that subassembly hardware was shipped from EBR-II in 1976 with the shipments going to the ANL-W RSWF for interim storage. Some of these RSWF shipments were temporary. Other indirect disposal paths were used for some of the EBR-II subassembly waste shipments.

waste stream designated as ANL-MOD-1H. This new waste stream contained only irradiated hardware. The LLW waste shipped for disposal in ANL-765-2H and ANL-785-1H were merged into a new LLW stream discussed below.

The other redefined waste streams (see ANL-MOD-2H through ANL-MOD-4H) that include irradiated fissile material consisted of driver fuels, experimental test fuels, irradiated flux wires, blanket material, reprocessing waste, and bulk shipments of low or unirradiated uranium. Much of the irradiated fissile material was generated from the destructive examination of highly enriched fuel elements. Generally, fuel samples were chemically dissolved and stabilized in an inert material like vermiculite; these shipments usually had actinide contents that weighed less than 200 g.

A major fraction of what was identified as dry active waste (mainly waste from hot cell operations including material like concentrated evaporator bottoms) disposed of during the 1960s had significant amounts of mixed fission products and actinides as byproducts of the EBR-II reprocessing campaign. In the previous waste stream breakdown, mainly hot cell waste was identified (see Table 3; i.e., waste streams ANL-752-1H, ANL-765-1H, and ANL-767-1H). However, the dry active waste definition is so general that a distinction between high-volume LLW and more concentrated fission-actinide contaminant disposals is unclear. Thus, high-activity waste stream components (e.g., irradiated fuel dissolved in vermiculite or irradiated flux wires) were made distinct from LLW general plant radioactive waste production. Since disposal and reporting practices varied between 1960 and 1983, two new waste streams containing high-fission product activities along with actinides were redefined as ANL-MOD-3H and ANL-MOD-2H over the years 1952–1970 and 1971–1983, respectively.

Bulk shipments of actinide waste usually contained unirradiated uranium (e.g., uranium oxide, natural uranium metal, and depleted uranium). Generally, these bulk shipments had actinide weights (usually uranium) exceeding 50 kg and originated from disposals of ceramic waste from ZPR3 and ZPPR, blanket waste from EBR-I, and smaller amounts of waste from unidentified sources. Because these shipments were sporadic, this waste stream was combined with two other special purpose waste streams, including the disposal of a radium source capsule and an irradiated tritium test capsule (ANL-MOD-4H). The newly defined "ANL-MOD-4H" waste stream includes elements of bulk shipments previously reported in other waste streams (e.g., as ANL-EBRI-1H).

All the solid, low-activity waste shipments (i.e., having no or only trace actinide contaminants) associated with waste streams ANL-752-1H, ANL-752-2H, ANL-752-3H, ANL-765-1H, ANL-765-1H, ANL-765-1H and ANL-EBRI-1H were reclassified as general plant waste and combined into waste stream ANL-MOD-5H.

Table 2. Original and current descriptions for ANL-W waste streams buried in the SDA during the years 1952 through 1983.

		Current Waste Stream Descriptions		
Waste Stream Number	Description of Waste	Waste Stream Number	Description of Waste	
	Dry active waste routinely generated from facility operations, maintenance, laboratory and sample waste.	ANL-MOD-1H	Irradiated subassembly hardware 1977 through 1983 for which principal contaminants were activation products.	
	Combustibles (paper, cloth, etc.); plastic, metal, and filters.	ANL-MOD-2H	Fuel-bearing waste such as unirradiated and irradiated fuel specimens 1971 through 1983. The principal contaminants were fission products and actinides. Extended data covers the period of 1984 through 1993.	
ANL-752-3H (Concentrated evaporator bottoms.	ANL-MOD-3H	Irradiated and unirradiated dissolved fuel, other miscellaneous irradiated fuel material, and fuel contaminated reprocessing hardware waste 1952 through 1970.	
	Dry active waste routinely generated in facility monitoring, operations, and maintenance activities.	ANL-MOD-4H	Bulk actinides and special purpose waste estimates ^b 1952 through 1983. The principal contaminants were uranium isotopes with smaller amounts of fission products in a small fraction of the disposals.	
	Subassembly hardware (from nuclear fuel and material experiments).	ANL-MOD-5H	General plant waste 1952 through 1983. These were high-volume LLW streams that had trace contamination of activation, fission, and actinide nuclides. Most streams were connected with decontamination operations.	
	Dry active waste routinely generated in facility monitoring, operations, and maintenance activities.			
	Subassembly hardware (from nuclear fuel and material experiments), rags, plastic sheeting, and equipment.			
ANL-EBRI-1H A	A wide range of waste from EBR-I.			

a. Original waste streams defined as part of the Historical Data Task (LMITCO 1995a).

b. Waste estimates calculated using an uncertainty factor of 2 (see Appendix A).

3.3 Data Collection and Analysis

The general approach to collecting data was to review any available documentation pertinent to SDA disposal operations. This documentation fell into two categories: (1) actual shipping manifests or electronic databases with information about SDA disposals and (2) documents with process information connected with actual disposals.^k

From 1960 through 1970, documentation was usually hard copies of standardized waste disposal Form 110¹. These forms generally reported waste types sent to the SDA along with net activities per shipment from 1960 through 1970. Most Form 110 data have been input into an electronic database called "WasteOScope."

For later disposals from 1971 through 1983, the INEEL RWMIS database provided a more detailed and reliable source of information—available on Excel spreadsheets—for estimating a complete nuclide-specific breakdown of COCs. These spreadsheets are chronologically ordered by year from 1971 through 1983. For each individual waste shipment, data entries included the date and location of the disposal, the general contents (waste stream identified numerically), and shipping container (identified numerically). These entries also had estimates of the net activities. Generally, the net activities also include other, more isotope-specific activities. However, the fission products reported by shipment are generally limited to activities for such contaminants as Cs-137 and Sr-90, without reporting other contaminants of interest such as Tc-99 and I-129. Form 110 data usually contained more detail information about the isotopic contents of each waste shipment. Information from the Form 110s and RWMIS for 1971–1983 was cross-checked for consistency.

Methods used to reassess SDA contaminant inventories for major identified waste streams are discussed in subsequent sections.

3.4 Overview of the Analysis Methodology

Table 3 summarizes the analysis methodologies that were used to analyze each waste stream. The location of the best-estimate inventories for each waste stream are also identified in this table. Additional details about the processes that were used to determine the inventories in each waste stream are discussed in later sections and in the appendices.

k. Relevant process related documents are listed in the references section of this report (Section 5).

^{1.} U. S. Atomic Energy standard Form 110, "Idaho Operations Office Waste Disposal Request and Authorization," (AEC 1964). In later periods, the shipping form number transitioned from 110 to 135. However, in this report 110 and 135 are considered as equivalent names.

Table 3. Summary of the analysis methods used to analyze each ANL-W waste stream for the HDT.

ANL-W Waste-Streams	Primary Waste Types	Analysis Methods	Location of Best-Estimate Results
ANL-MOD-1H	Irradiated SA hardware	~1,800 SA were identified as being disposed at the SDA. The estimated inventories of activation products were associated with each SA based on its EBR-II core position and benchmark inventories. See Appendix C for analysis details.	Table B-18 and Tables C-2, C-3
ANL-MOD-2H and ANL-MOD-3H	Fuel-bearing waste including irradiated and unirradiated dissolved fuel and fuel contaminated materials	Scaling factors were used, along with the estimated mass of heavy metals (from Appendix A) that were disposed at the SDA to estimate the inventory of fission products and actinides that should be presented in these wastes.	Tables B-12 and B-9
ANL-MOD-4H	Low or unirradiated bulk-actinide waste	The principal contaminants consisted of uranium isotopes and a few other actinides like Pu-239 and Np-237. Inventories were assessed based on the reported shipping data. Except for an estimate of the U-234 inventory (based on the quantity of U-235), no other calculations were made to determine fission product or actinide inventories due to the very low irradiations that these wastes experienced.	Table B-1
ANL-MOD-5H	General plant wastes consisting of small amount of fission products, actinides, and activation products	Scaling factors were used along with the reported Cs-137 activities to determine the inventory of fission products, actinides, and activation products that are probably present in these waste items.	Table B-24
SA = subassembly			

3.5 Activation Product Inventory

Irradiated subassembly hardware were shipped from EBR-II to the SDA soil vaults from 1977 through 1993.^m Estimates of activation products recorded in the RWMIS database did not include all required nuclides needed for a risk analysis (e.g., Tc-99 or Ni-59). Consequently, supplemental calculations fill known gaps in the inventory of activation-products in the HDT and RWMIS database. Typical burnup conditions for EBR-II driver fuel rods were employed in a series of reactor physics simulations to calculate the buildup of activation products. These simulations generated approximate activation-product activity levels for subassembly components sent to the RWMC.

m. After 1993, irradiated hardware was disposed of in concrete-lined silos at the RWMC.

The EBR-II core comprised a hexagonal lattice of various subassembly types. Several generations of fuel designs (e.g., Mark-I, Mark-IA, Mark-II, and Mark-III) were used in EBR-II, and the burnups varied considerably over individual recorded histories. Subcategories of these different components included such subassembly classifications as core or driver fuels and inner and outer blanket. These defueled subassembly components usually included top and bottom reflector pieces and a cylindrical, hexagonal outer shell that surrounded the fuel pins. In addition, irradiated control rod assemblies also were included in the waste streams sent to the RWMC. The materials composition of the subassembly components was either 304 or 316 stainless steel. The best-estimate HDT inventories are shown in Table 4 and were taken from more detailed tables that appear in Appendix B.

Table 4. Comparison of activation-product activities for the years 1952 through 1983.

Nuclide	RWMIS Reported Total Activity (Ci)	HDT Reported Total Activity (Ci)	Best-Estimate Activity ^a (Ci)
C-14	_	_	1.61E+01
Cl-36	_	_	4.00E-03
Co-60	5.10E+05	3.30E+05	8.09E+05
Ni-59	_	_	8.57E+01
Ni-63	_	_	6.27E+03
Nb-94	_	_	2.81E+00
Tc-99		_	$6.88E+00^{b}$

HDT = Historical Data Task (LMITCO 1995a)

RWMIS = Radioactive Waste Management Information System

Irradiated subassembly hardware sent to the SDA from 1977 through 1983 contained almost all of the activation products identified during the HDT and RPDT periods. Records from the RWMIS show hundreds of waste shipments with small amounts of activation-product contaminants were sent to the SDA pits. However, the net activated-product pit disposals were less than 3% of the activity contained within the subassembly hardware from EBR-II. As in later shipments made during the RPDT period, the subassembly components consisted of highly irradiated 304 and 316 stainless steel alloys. The total reported activities from the RWMIS database and HDT are also shown in Table 4. These data are compared against the best-estimate results from the current reassessment analysis (obtained from Table B-18 in Appendix B).

a. Obtained from Table B-18.

b. This value represents the Tc-99 inventory in subassembly hardware only. A contribution also exists in fission product contaminated wastes.

3.6 Estimated Actinide Inventory

Disposals of both irradiated and unirradiated actinide (e.g., heavy metal) wastes were identified in shipping records from 1960 through 1988. As previously mentioned, no data were found regarding ANL-W waste shipments prior to 1960. The radioactive waste streams that were identified included dissolved (irradiated and unirradiated) fuel samples and bulk-actinide disposals of both depleted and natural uranium. In general, these waste streams fall into two categories: (1) sporadic shipments generally having significant bulk weights of unirradiated or low-irradiated actinide waste (mainly uranium); and (2) highly irradiated fuel samples or irradiated test specimens with weights that were generally less than 100 g (not including the container weight). However, some shipments contained unirradiated fuel specimens as well.

The majority of the waste in the second category was shipped in heavily shielded stainless steel containers. The first category includes bulk uranium-based waste streams that were generally low-enriched or depleted uranium that were either unirradiated or had extremely low irradiation histories. Most of this waste was shipped in unshielded containers, including wooden boxes and thin-walled metal containers. The amounts of fission-product contaminants were generally low or nonexistent for these waste streams. These conclusions follow from the low gross activities for most of the disposals of bulk-actinide waste. However, this category also includes neutron-irradiated blanket material from EBR-I containing some TRU contaminants that resulted from the breeding of plutonium (Haroldsen et al. 1961; Smith et al. 1961). See Table 5 for a summary of major actinide waste disposals for 1960 through 1983. Based on the total weights shown in Table 5, the individual TRU isotopes were determined. For example, all of the thorium was assumed to be Th-232 and all of the plutonium was assumed to be Pu-239. The uranium mass data was separated into U-235 and U-238 isotopes, and then converted into curies. The U-234 inventory was determined based on expected ratios with U-235, depending on whether the source of uranium was natural, depleted, or enriched. The best-estimate activity data corresponding to the reported mass data from Table 5 is shown in Table B-1 of Appendix B.

From Table 5, a total of 3,810 kg of bulk-actinide waste was disposed of in the SDA from 1960 through 1983. Individual bulk waste streams include ceramic oxide waste originating from the ZPPR and ZPR3 facilities. Table 5 also shows some thorium, highly enriched uranium, and plutonium designated "bulk special-purpose waste" whose origins are unknown; nevertheless, they were considered to come from ANL-W facilities. Some of the depleted uranium shipments were neutron irradiated. One depleted uranium disposal shipment made in 1969 was identified as a 95-kg ingot of depleted uranium 5% fissium material." Melt refining of irradiated EBR-II blanket rods most likely produced the depleted ingot with a reported activity of less than 1 Ci. During the casting process, most of the fission products were removed and a low-activity metal ingot with TRU contaminants was produced. Other reported bulk shipments may have been shielding or reflector materials.

The second category of explicitly reported actinide waste streams included unirradiated or irradiated driver fuel, test specimens, and a variety of special-purpose wastes that were generally dissolved in vermiculite or other stabilizing media. The corresponding irradiation histories of these waste streams are unknown. Typically, EBR-II driver fuel or test samples were chemically dissolved for laboratory assays and then mixed with vermiculite for disposal.

Several subcategories of special-purpose waste were sent to the SDA. One subcategory included irradiated fission detectors and flux wires, which were generally composed of enriched uranium. Another subcategory included special irradiation test capsules in 1981 (e.g., test capsules containing 6.2 g of neptunium-237). Irradiated driver fuel or test samples were also byproducts of reprocessing operations in the 1960s. A few shipments of broken glass molds contaminated with processed fuel were sent to the

n. The EBR-II uranium fuel was alloyed with other metals (e.g., molybdenum) at approximately 5% fractional weight.

o. These TRU contaminants resulted from neutron capture by uranium atoms.

SDA. Other miscellaneous shipments of reprocessing waste containing EBR-II fuel, actinide waste, and fission-product contamination were also reported. A number of shipments were classified as hot cell dry active waste but were mixed with reported quantities of driver or sample fuel contaminants. The Form 110sfrom the 1960s indicate disposal in the SDA of fuel and irradiated test samples from EBR-I and BORAX (both located outside the boundaries of the ANL-W site). This suggests that irradiated fuel samples from these two programs were also sent to the SDA during the 1950s. If this is correct, then these waste shipments could be classified under "ANL-E" or "Other" facilities; and therefore, removed from the ANL-W listing. However, since some of these wastes may have traveled through ANL-W (at least the shipping records), they were included in this reassessment. One shipping manifest recorded irradiated EBR-I material consisting of 125 blanket rods of depleted uranium. However, other documents suggest that 25 of these rods were possibly irradiated plutonium driver fuel (Kendall and Wang 1975). These rods are shown as 2.4 kg of Pu-239 for 1964 in Table A-5 of Appendix A.

Data from the Form 110 shipping manifests and records of ANL-W remote-handled waste are summarized in Appendix A. These data were used to calculate the mass of bulk-actinide and fuel-bearing wastes (e.g., heavy metals) disposed at the SDA. This information is summarized in Table 5. In one case, historical documents were used to supplement a Form 110 shipping manifest to estimate the actinide inventory in a disposal of EBR-I fuel rods that was made in 1964 (see Table A-5 in Appendix A).

-

p. During this period, waste containing highly enriched uranium was reprocessed at INTEC.

q. By cross-checking the shipping manifest description of the zirconium-clad depleted rods with EBR-I reports, the INEEL concluded that only Core IV matched the physical description of the depleted rods. One ANL-W technical report description (Haroldsen et al. 1961) shows that the other inner blanket rod material was stainless steel-clad natural uranium. Report data also show for Core IV that only 100 depleted rods were incorporated into the plutonium experimental core region. This observation suggests that 25 rods were actually plutonium driver fuel rods. The high activities in this 1964 shipment also suggest that plutonium driver fuel was present.

Table 5. Best-estimate mass inventory of bulk-actinide wastes (heavy metals) with little or no irradiation that were sent to the SDA from 1960 through 1983^a, and are included in waste-stream ANL-MOD-4H.

SDA Disposal Year	EBR-I Blanket (kg)	Depleted Uranium (kg)	Uranium- Oxide Ceramic (kg)	Natural Uranium (kg)	Enriched Uranium (kg)	Total Uranium (kg)	Plutonium (kg)	Neptunium (kg)	Thorium (kg)
1960	_		_	_	_	_	_	_	_
1961	_		_	_	_	_	_	_	_
1962	_	_	_	_	_	_	_	_	_
1963	_	_	_	_	_	_	_	_	_
1964	3.10E+01	_	_	_	_	3.10E+01	_	_	_
1965	8.88E+02	_	_	_	_	8.88E+02	_	_	_
1966	_	_	_	_	_	_	_	_	_
1967	4.57E+01	8.34E+01	_	1.31E+01	1.75E+00	1.44E+02	3.15E-01	_	1.06E+00
1968	_	6.18E+00	_	1.33E+01	1.78E+00	2.12E+01	5.40E-01	_	1.08E+00
1969	_	9.50E+01	7.23E+02	_	_	8.18E+02	_	_	_
1970	_	_	1.85E+02	3.31E-01	_	1.85E+02	_	_	_
1971	_	_	_	_	_	_	_	_	_
1972	_	_	1.30E+02	_	_	1.30E+02	_	_	_
1973	_	_	_	_	_	_	_	_	_
1974	_	_	_	_	_	_	_	_	_
1975	_	_	_	_	_	_	_	_	_
1976	_	3.18E+02	1.27E+02	_	_	4.45E+02	_	_	_
1977	_	3.12E+01	5.60E+01	_	_	8.72E+01	_	_	_
1978	_	7.20E+01	1.91E+02	_	_	2.63E+02	_	_	_
1979	_	3.60E+01	4.30E+02	_	_	4.66E+02	_	_	1.00E-02
1980	_	2.70E+01	2.00E-02	_	_	2.70E+01	_	_	_
1981	_	_	_	_	_	_	_	6.2E-03	_
1982	_	1.65E+02	_	_	_	1.65E+02	_	_	_
1983	_	2.50E+01	1.12E+02			1.37E+02			
Total	9.65E+02	8.59E+02	1.95E+03	2.67E+01	3.53E+00	3.81E+03	8.55E-01	6.2E-03	2.15E+00

Total mass of all low irradiated heavy metals (i.e., uranium) = 3,810 kg (this material was included in the ANL-MOD-4H waste-stream).

EBR = Experimental Breeder Reactor

SDA = Subsurface Disposal Area a. No data exists for 1952-1959. Therefore, activities shown in this table also apply for 1952 through 1993.

Because the irradiation histories associated with bulk-actinide wastes in Table 5 were very low, fission product and TRU isotopes resulting from fission or capture events were considered negligible for waste stream ANL-MOD-4H (1952 through 1983). Therefore, for the bulk-actinide wastes, no fission products or TRU isotopes (beyond those isotopes already reported by the waste generator) were estimated to exist with these waste streams (see Table B-1). However, in the case of fuel-bearing waste shipments, as illustrated in Table 6 (obtained from Appendix A), these wastes usually consisted of irradiated fuel specimens or similar material (e.g., irradiated flux wires, etc.). In this case, calculations were made (based on scaling factors) to predict the inventory of fission products and TRU that probably were present in these heavy-metal wastes, but were generally not reported. These calculations were applied to waste streams ANL-MOD-3H and ANL-MOD-2H, but not to waste streams ANL-MOD-4H (or ANL-MOD-2R). These calculations are discussed in greater detail in Section B-3 of Appendix B.

Table 6. Shipping data for irradiated fuel-bearing wastes (e.g., fuel samples) sent to the SDA from 1960 through 1983^a (heavy metals mass data included in ANL-MOD-3H and ANL-MOD-2H).

Disposal	Net Actinide or Heavy Metals Mass Disposed at the SDA ^b	Reported Activity that was Shipped ^c
Year	(kg)	(Ci)
1960	1.00E-05	3.0E-04
1961	6.03E-02	1.0E-01
1962	8.30E-04	6.5E+00
1963	1.75E-01	5.4E-03
1964	2.69E+00	1.6E+02
1965	2.87E-02	3.2E+02
1966	4.30E-01	2.1E+03
1967	2.32E+00	5.1E+03
1968	5.41E+00	1.7E+04
1969	3.91E+00	3.2E+03
1970	2.30E+00	1.9E+02
1971	4.61E+00	2.9E+03
1972	5.20E+00	6.9E-03
1973	1.66E+00	5.5E-01
1974	5.22E-01	3.3E+02
1975	2.55E+00	2.4E+01
1976	1.93E+00	6.5E+00
1977	1.13E+00	7.3E+01
1978	4.72E-01	3.7E+02
1979	9.02E-02	5.1E+00
1980	2.33E-01	1.3E+01
1981	3.45E-01	1.2E+01
1982	<u> </u>	_
1983	8.61E-01	1.2E-01
Total	3.69E+01	3.2E+04

a. No data exists for the period 1952 through 1959.

The actinide data corresponding to the detailed analysis presented in Appendix B are summarized in Table 7 for the period 1960 through 1983. These data are also compared with similar data from the corresponding HDT report (LMITCO 1995a). Note that the inventories of uranium isotopes are in

b. Obtained from the underlined "total" values shown in Tables A-1 through A-24 of Appendix A.

c. The activity data were reported on the Form 110-shipping manifests and may not correlate with curies calculated from the individual radionuclides. See Appendix A for a detailed list of radionuclides.

reasonable agreement (within a factor of 3). However, the amounts of Th-232 and Pu-239 are significantly higher than the corresponding HDT inventory values. Differences in the Pu-239 activities appear to originate from the inclusion of new data based on Form 110 shipping records (i.e., three shipments of low-irradiated actinide waste) and from the reasonable evidence that 25 irradiated plutonium driver rods from EBR-I were disposed of in the SDA (see the 1964 disposal data for Pu-239 as listed in Table A-5). Discrepancies in the inventories of Th-232 may be the result of the better availability of historical shipping records for this report. Finally, there are no data for Np-237 in the HDT (LMITCO 1995a) or RPDT (LMITCO 1995b) to compare against.

Table 7. Best-estimate actinide disposals made at the SDA from 1960^a through 1983.

Nuclide	Best-estimate Inventory ^a for Bulk Actinides (Ci)	HDT Inventory for Bulk Actinides (Ci)
Th-232	8.49E-04	1.0E-05
U-234	2.16E+00	3.4E+00
U-235	9.18E-02	2.7E-01
U-238	1.30E+00	1.2E+00
Pu-239	$4.39E+02^{b}$	1.1E+01
Np-237	2.32E-02	_

HDT = Historical Data Task

Similar to the calculation of fission products present in general plant wastes, due to the presence of Cs-137; actinides are expected to exist in waste streams that contain Cs-137 (or those waste streams that contain irradiated heavy metals). In other words, Cs-137 is probably present because it is associated with irradiated fuel particles; therefore, fission products, uranium, and actinides should also be present. Details regarding the estimated inventory of actinides present in waste streams associated with Cs-137 can be found in Appendix B (see Tables B-9 B-12, B-15, and B-24).

The presence of unirradiated actinide contaminants is separately factored into the total inventory. In addition, this method may not be directly applicable to contaminants from specialized irradiation experiments that have irradiation histories and compositions that deviate significantly from standard EBR-II driver fuel characteristics.

Explicitly identified waste streams with actinide contaminants (amounts reported by weight) from remote shipping records are thought to be irradiated fuel samples. However, the bulk of the actinide waste came from one shipment. In that shipment, approximately 16 kg of uranium was disposed of in the SDA soil vaults. The associated irradiation history of this disposal and the source of the uranium are of unknown origin. In a number of cases, irradiated subassembly material was mixed with an identified actinide waste stream in the same shipping container. The actinide waste material could be samples of EBR-II driver fuel, irradiated test fuel specimens, or materials from other facilities. Until more information becomes available, it is assumed that this actinide waste was from EBR-II operations. Although INEEL personnel have physical possession of many internal remote-handled waste-shipping records for the RPDT period, it is not clear that all documents are included. The RWMIS inventories of

24

a. No data exists for the period 1952-1959. Since disposals from ANL-W to the SDA would have been small during this period, then these results can also be applied to 1952 through 1983.

b. Some records indicate that a shipment of 2.4 kg (149 Ci) of plutonium contained within 25 EBR-I fuel rods was disposed at the SDA in 1964.

r. Most of the Form 110 records used in the reassessment are now electronically available from the INEEL "Environmental Restoration Optical Imaging System Retrieval").

actinide waste streams showed that about 250 kg of unirradiated uranium (either depleted or natural) was sent to the SDA. Additional information shows that this material was low-irradiated ceramic uranium oxide blanket material from the ZPPR facility^s. Generally, shipment-specific enrichments and chemical forms are unknown. Waste descriptions for RWMIS pit shipments for 1984–1993 were encoded with generic identification numbers. The RWMIS database did not always identify the process that generated these actinide waste streams or associated disposed material states (e.g., depleted uranium, natural uranium, and oxide).

3.7 Estimated Fission Product Inventory

The review of fission product contaminant disposals can be separated into four different time intervals: (1) 1952 through 1959 (essentially no waste was disposed at the SDA from ANL-W during this time), (2) 1960 through 1970, (3) 1971 through 1983, and (4) 1984 through 1993. The HDT period covers 1952 through 1984, and the RPDT covers the period 1984 through 1993 (and is discussed in Section 4.7) As previously mentioned, no ANL-W data exists for 1952-1959. Since most of the ANL-W facilities were not operating during this period, it is believed that very little radioactive waste was sent to the SDA prior to 1960. From 1960 through 1970, only gross activities appear in the RWMIS database. In addition to the RWMIS data, gross activity data has been reported by Witbeck and Fryer (1979) (for 1968 through 1978) and also in Form 110 shipping records. From 1971 through 1983, contaminants were broken down into separate categories (e.g., fission products, actinide waste, or activation products). Detailed isotopic information from the RWMIS database begins in 1971; however, the Form 110 shipping data begins around 1960.

Exact burnup histories for irradiated fuel and test specimens are unknown. Fission byproducts from irradiated fuel and/or fuel specimens were the main contributors to fission-product activities with a sizeable fraction originating from the HFEF. Part of these contaminants originated from hot cell operations with associated fission byproducts being generically classified as dry active waste. Decontamination activities that produced evaporator waste represent a smaller waste stream of secondary fission products. These two streams were generally free or had low amounts of secondary actinide contamination. Irradiated fuel-based waste streams had attendant actinide contaminants from neutron captures that generated Pu-239, Pu-240, Np-237, and Am-241. TRU isotopic contaminants were not generally broken out in detail for this period. To complicate matters, a sizable fraction of the net activity was either designated as mixed fission products (MFPs) or unidentified alpha (UN-ID-Alpha) and unidentified beta+gamma emitters (UN-ID-B+G). Most fission-product activity disposed of from 1971 through 1983 was produced from hot cell operations.

Destructive examination of fuel or test specimens generated multiple waste streams, including waste production designated as dry active waste, dissolved fuel specimens, filter, and evaporator waste streams. The evaporator system concentrated both liquid hot cell decontamination solutions and several secondary waste streams that included resins, various filters, and evaporator bottoms (Benedict, Pigford, and Levi 1981).

Based on Table 1, from 1960 through 1970, a gross activity of ~49,000 Ci was buried in the SDA. Most of the disposal activity was associated with the EBR-II fuel-reprocessing program that was begun in the 1960s. Three dominant waste streams containing fission product waste were associated with reprocessing actions. Destructive testing of irradiated fuel resulted in numerous disposal shipments of dissolved fuel that was usually stabilized in vermiculite. Contaminated reprocessing equipment was also a major waste stream (e.g., disposals of melt refining induction furnaces). In addition, routine hot cell cleanup operations generated significant amounts of dry active waste. Shielded packaging of these three

25

s. R. P. Grant (ANL-W) to N. Hampton (BBWI), April 3, 2003, "Development of a Comprehensive Inventory of ANL-W Radiological Contaminants Buried in the Subsurface Disposal Area of the INEEL RWMC During the Period 1952 Through 1993."

waste streams was generally done before transport to the SDA. Because of high gamma fields, most of this waste was buried in trenches rather than pits.

It is probable that from 1952 through 1959, similar kinds of wastes were generated from routine operations at EBR-I and the BORAX facilities. Although no records from this period have been located, it is expected that the total amount of wastes from these facilities (from 1952 through 1959) was relatively small compared with the wastes generated from ANL-W facilities after 1960. This conclusion is consistent with the following notation mentioned in the HDT report: "The EBR-I, ZPR-3 and Borax facilities and waste records available at ANL-W show no disposal at RWMC until 1958."

Fission product inventories were estimated to be present in the following HDT fuel-bearing waste streams: ANL-MOD-3H and ANL-MOD-2H, and in the general plant waste stream: ANL-MOD-5H. Fission products were estimated to be present in fuel-bearing wastes because of the heavy-metal mass inventories (usually uranium) and the fact that these wastes generally originated from irradiated fuel. Fission products were estimated to be present in general plant wastes due to the presence of Cs-137. The estimated inventories of fission products in these two categories was accomplished in similar ways. For the fuel-bearing wastes, the inventory of unreported fission products (or activation products) was estimated by multiplying scaling factors (Ci/kg-HM) by the reported heavy metal inventory (kg). For general plant wastes, the inventory of unreported radionuclides was estimated by multiplying scaling factors based on Cs-137 (Ci/Ci-Cs137) by the estimated Cs-137 activity (Ci) in this waste. The scaling factors used in either of the two categories are related by the following relationship:

- Scaling Factor (Ci/Ci-Cs137) = [Scaling Factor (Ci/kg-HM)] / [Cs137 scaling factor (Ci-Cs137/kg-HM)]
- See Section B-3 in Appendix B for additional details concerning the calculation and use of scaling factors, and the estimated inventories in these waste streams.

Estimates of many fission products (except Cs-137) were calculated from scaling factor tables." That is, an average isotopic profile was used to quantify the activities for the HDT and RPDT periods. A single gamma emitter was selected from which all other radionuclides could be determined. In this report, Cs-137 was chosen as the reference isotope because it can be easily detected and was consistently reported in many RWMIS shipments. Logged Cs-137 activities constituted a significant fraction of the total reported MFP activities. Net Cs-137 gamma activities were then scaled to calculate the activity of other nuclides of interest—such as I-129—that were not explicitly reported.

The scaling methods used in this reassessment study are based on highly irradiated EBR-II driver fuel. Representative scaling factors for a typical EBR-II driver fuel element are listed in Appendix B (see Table B-8). Again, the derived scaling factors are referenced to Cs-137, and are ultimately based on reactor physics calculations of the inventory present in EBR-II driver fuel pins at maximum burnup conditions (see Section B-3 of Appendix B for additional details). Waste associated with experimental fuel assemblies would have been generated during destructive examination procedures. The isotopic

t. These shielded containers were emptied rather than buried with the waste disposed of in the SDA.

u. A significant fraction of the disposed fission products and TRU contaminants were connected with EBR-II operations: processes for handling irradiated fuel and decontamination. These process waste streams would have included disposals of LLW connected with hot cell operation and decontamination. Other minor waste streams containing fission products contaminants originated from TREAT, ZPPR, and other support facilities. Contributions from support facilities were typically not as significant as from EBR-II.

v. This mode of contaminant reporting was applicable only to waste streams contaminated with fission products. In addition, isotopic breakdowns of reported fission product contaminants were not always broken out. In many older shipping manifests, only net activities for the total amount of MFP were reported.

w. Examples of destructive examination include chemical dissolution of irradiated fuel or dissection of fuel in hot cells.

profile associated with experimental assemblies or other irradiated fuel capsules may differ from the profile from EBR-II driver fuel. Consequently, some uncertainties are introduced by applying one set of scaling factors to all waste shipments. However, since the vast majority of all fuel that processed (or reprocessed) at ANL-W facilities was from EBR-II driver fuel assemblies, this assumption is probably reasonable.

Waste streams containing fission products (and actinides) were buried in both pits and soil vaults. A detailed yearly breakdown of activities disposed at the SDA is shown in Appendix B. Activity totals are summarized in Table 8 for selected fission products and were obtained from Table B-31 (Part 5). This data is also compared against information from the HDT (LMITCO 1995 a).

Table 8. Summary of the total fission product waste activities that were disposed at the SDA from ANL-W operations from 1960 through 1983.^a

Nuclide (Ci)	Best-estimate Activity (Ci)	HDT Activity (Ci)
H-3	1.28E+02	_
Sr-90	1.69E+04	2.2E+05
Tc-99	$9.74E+00^{b}$	_
I-129	5.90E-03	_
Cs-137	2.34E+04	1.4E+05
Eu-152	1.19E+00	
Eu-154	1.24E+02	
Total	4.04E+04	3.6E+05

a. No data exists for the period 1952-1959.

b. Tc-99 appears as both a fission product and as an activation product. The listed number includes both contributions; however, the activation component is identified as 6.88 Ci for waste stream ANL-MOD-1H in Table B-18.

Numerical totals were directly calculated from Excel spreadsheets. These spreadsheets were built shipment-by-shipment using RWMIS data. Activities for some COCs were not reported at the time of disposal. Typically, the missing entries were the consequence of reporting criteria used at that time, including gross MFP activities reported without any additional isotopic breakdown for a particular shipment. To estimate other unreported nuclides of interest, scaling factors from Appendix B were proportioned to the Cs-137 activities sent to the SDA.

However, this scaling approximation assumes that most of the ANL-W waste streams were contaminated with EBR-II driver fuel material. That is, fuel contaminated wastes from TREAT and other ANL-W facilities was small compared with waste streams that originated from EBR-II operations, and this assumption appears to be correct.

3.8 Uncertainties

Because of the highly variable and shipment-dependent nature of many of the waste streams, standard statistical uncertainty methods were not feasible to define individual uncertainty factors. As in the original HDT and RPDT reports (LMITCO 1995a, 1995b), the methodology for defining the best-estimate activities and associated upper and lower bounds was not based on a rigorous statistical error propagation model. Instead, approximation methods were based on professional (e.g., engineering) judgment and other reasonable assumptions.

Estimates of contaminants for waste streams containing activation products, fission products, and actinide products are all affected by incomplete documentation as well as other factors needed to make estimates having reasonable uncertainties. The major contributors that affect uncertainties values are:

- Uncertainties in reporting the irradiation history and composition of materials
- Uncertainties due to undocumented burials or disposals of materials
- Uncertainties due to computer code calculations or calculations that used scaling factors (caused by cross-section uncertainties, decay times, numerical solution of the differential equations, etc.).
- Measurement or reporting uncertainties of key isotopes (e.g., Cs-137).x
- Interpreting the shipping records for the period to correlate a waste stream process with the disposal manifest of a specific shipment.

Based on engineering judgment, an uncertainty factor of <u>at least</u> 1.5 should be applied to the estimated activity of most radionuclides. However, in some specific cases, uncertainty factors might be as high as a factor of 100. In the case of the bulk-actinide waste stream ANL-MOD-4H, an uncertainty factor of 1.50 was assumed for all radionuclides. That is, for these waste streams, the Upper-bound activities were estimated by multiplying the best-estimate activities by a factor of 1.50; and the Lower-bound activities were estimated by dividing the best-estimate activities by 1.50. See Tables B-1 through B-5 for additional details.

In the case of waste streams activities that were estimated with the aid of scaling factors (e.g., ANL-MOD-3H), then individual uncertainty factors were applied to each radionuclide. These factors ranged from 1.1 for U-235 up to a factor of 100 for Cl-36. Additional details are shown in Table B-7 of Appendix B.

x. Some indirect measurement techniques for high activity shipments (e.g., Co-60) had errors that underestimated disposals by as much as a factor of ten.

As a simple example of the creation of a waste stream and record at that time, consider the destructive examination in a hot cell of fuel elements from EBR-II at HFEF. After the examination, the hot cell equipment was decontaminated. The fission-bearing byproducts of decontamination are subsequently processed and sent to the SDA. The corresponding shipping manifest would identify the building and container numbers from where the materials originated, but would not describe the irradiation history connected with the attendant waste fission products.

Thus, assumptions were made concerning irradiation histories of the associated waste stream to provide bounding estimates of ANL-W contaminant inventories disposed of in the SDA. Items from the above list further complicated efforts to precisely define attendant uncertainties. Because of these difficulties, the same uncertainty factors for the HDT were often applied to the RPDT period (see Section 4.8).

Limited disposal information proved to be a major contributor to the above-mentioned uncertainties. Associated estimates of reassessed contaminants presented in Appendix B have significant uncertainties for the following reasons:

- A search at the INEEL has failed to locate most of the Form 110 soil vault disposal records from 1977 to 1983. Locating the missing Form 110s for the soil vaults will help resolve whether any additional actinide-based waste streams were sent to the SDA soil vaults.
- During the 1950s, disposals of dissolved fuels and test samples from BORAX, EBR-I, and other
 unknown irradiated actinide waste materials may have been sent to the SDA. In any case, it is not
 clear if disposals made from these facilities should be included under the ANL-W heading, or
 classified as originating from ANL-E or from "Other facilities," as noted in the HDT (LMITCO
 1995a).
- The Form 110 shipping manifests are incomplete for actinide-bearing waste streams.
- Disposal histories associated with EBR-I operations have many gaps, including the lack of disposal information for substantial amounts of irradiated actinide waste material from the EBR-I complex. These actinide waste streams include driver fuel, blanket material, and reflector material. Form 110 shipping manifests document the disposals of significant amounts of what is clearly EBR-I irradiated blanket material in the SDA. In addition, disposition records for all of the EBR-I Core IV plutonium driver fuel have not been located. Also, it is likely that destructive testing of EBR-I fuel resulted in some disposals of miscellaneous fuel samples in the SDA. From examination of INTEC fuel reprocessing records, it appears that most of the highly enriched driver fuels from the first three EBR-I cores were reprocessed at INTEC and not sent to the SDA.
- It is possible that most of the EBR-I Core IV plutonium fuel and blanket material may have been sent to the Radioactive Scrap and Waste Facility or reprocessed. The amount of incompletely documented irradiated blanket or fuel material presented in Table 9 is based on ANL-W EBR-I technical reports (Haroldsen et al. 1961; Smith et al. 1961; Kendall and Wang 1975).

Table 9. Irradiated blanket and fuel material from EBR-I for which disposition histories are incomplete or unknown.

	Mass of Incompletely
Item	Documented Material

y. Most of the inventory of contaminants buried in the soil vaults listed in this report are based on data taken from the electronic RWMIS database.

z. In addition, some of this material may have been sent off-Site for reprocessing (e.g., Hanford facility).

	(kg)	
Natural uranium inner blanket rods	2.9E+02	
Core I to III axial natural uranium blanket Material	2.5E+02	
Core II driver fuel	3.0E+00	
Core IV axial depleted uranium slugs above and below the driver fuel in same fuel element	3.8E+01	
Core IV Pu-239 driver fuel	2.8E+01	
Total	6.1E+02	

Disposals of processed radioactive NaK EBR-I coolant also were sent to the SDA as part of the DD&D operations. About 5,500 gal of EBR-I NaK coolant were processed and stored at Central Facilities Area before disposal to the SDA. This waste stream was judged as a minor component of the total EBR-I contaminant generation. A much smaller amount of NaK associated with the EBR-I 1955 meltdown event was disposed of after 1993 and did not fall directly within the scope of this document. ^{3a} Contaminants associated with the NaK included both fission products and metallic fuel fragments associated with fuel leakage or breaching.

The facility located next to EBR-I, ZPR3, generated waste containing actinide contaminants. Both facilities were subjected to complete or partial DD&D actions. The ZPR3 was also subjected to DD&D (Brown et al. 1987; Kittel, Novick, and Buchanan 1957; Little et al. 2001). However, technical data on ZPR3 was limited; consequently there is a significant uncertainty concerning actinide disposals associated with this facility. Waste streams from ZPR3 may have been recycled, reprocessed, or sent to the SDA. Disposals of some ceramic uranium oxide waste from ZPR3 were sent to the SDA during the HDT period (primarily in 1969).

aa. The NaK associated with the EBR-I Core II meltdown event was stored for many decades in a shielded bunker and was finally treated at the Sodium Components Maintenance Shop (SCMS). Final disposal in the SDA was accomplished after 1993. The associated isotopics related to this disposal were documented in Little et al. (2001).

4. ESTIMATED RADIOLOGICAL INVENTORY FOR 1984 THROUGH 1993

This section discusses the reassessment of waste sent from ANL-W to the SDA for disposals that occurred during the RPDT period from 1984 through 1993. The three main radiological contaminant categories associated with this period were fission products, activation products, and actinide contaminants.

4.1 Background

During the years 1984 through 1993, yearly activity disposals to the SDA exhibited variable trends ranging from 1.2E+04 to 4.6E+05 Ci, with net disposals amounting to 1.5E+06 Ci. These disposal trends are quantified in Table 10, showing yearly inventory disposals of net radionuclide activities. Activity trends are controlled mainly by the disposals of subassembly hardware. Only a small fraction of the yearly net activity disposals was attributed to fission product or actinide contaminants.

Waste generated at ANL-W from 1984 through 1993 consisted of both (1) contact-handled waste (low-activity waste that did not require shielding precautions; and contained varying small amounts of fission, activation, and actinide contaminants), and (2) remote-handled waste (high gamma fields that required safety precautions; e.g., transport in shielded containers). These two classes of waste were disposed of differently in the SDA. During the RPDT period, remote-handled waste was buried in soil vaults while contact-handled waste was buried in much larger pits. Burial in soil vaults ensured a higher level of shielding protection during remote waste handling. The relative net activity from the streams of contact-handled waste was a small fraction of the remote-handled waste activity.

Table 10. Summary of gross activity by year of waste shipped from ANL-W and disposed of at the SDA from 1984 through 1993.

Year	Gross Activity (Ci)	Important Events
1984	1.2E+05	Disposal practices during the years 1984–1993 were relatively uniform. EBR-II produces 13,600 MW _t D.
1985	1.3E+05	EBR-II produces 15,200 MW _t D.
1986	5.7E+04	EBR-II produces 14,600 MW _t D.
1987	2.0E+05	EBR-II produces 16,700 MW _t D.
1988	1.4E+05	EBR-II produces 17,900 MW _t D.
1989	4.6E+05	EBR-II produces 8,200 MW _t D.
1990	1.3E+05	EBR-II produces 23,400 MW _t D.
1991	8.2E+04	EBR-II produces 5,500 MW _t D.
1992	9.3E+04	EBR-II produces 12,300 MW _t D.
1993	8.7E+04	EBR-II produces 8,900 MW _t D.
1994	1.2E+04	EBR-II produces 7,600 MW $_{t}$ D. EBR-II is officially shut down on September 30, 1994.
Total	1.5E+06	

4.2 Disposal Waste Streams

The original RPDT waste streams (LMITCO 1995b), and modified definitions of waste streams for the years 1984-1993 are shown in Table 11. The principal contaminants fall into three categories: activation products, fission products, and actinides (including TRU isotopes).

The original (LMITCO 1995a) and modified waste streams for 1984–1993 are presented in Table 11. Generally, the reassessment of data for this period indicated that original waste streams included in Table 11 were reasonably complete. However, to better profile these waste streams, descriptions were redefined using methods similar to those discussed above for the HDT.

The high-activity disposals of irradiated subassembly hardware previously identified as contained in waste streams ANL-765-2R and ANL-785-1R were combined into a single waste stream designated as ANL-MOD-1R. This new waste stream contained only irradiated hardware. Suspected or known irradiated fissile material disposed of as remote-handled waste was redefined as ANL-MOD-3R.

All of the low-activity, contact-handled waste shipments with trace or no actinide contaminants were reclassified and combined into waste stream ANL-MOD-4R. This stream includes low-level waste shipments from streams ANL-704-1R, ANL-752-1R, ANL-765-1R, ANL-785-2R, ANL-793-1R, and ANL-798-1R. Contact-handled bulk actinide waste streams such as ANL-793-1R were recombined into a new stream called ANL-MOD-2R.

4.3 Data Collection and Analysis

The general approach to collecting data was to review any available documentation pertinent to SDA disposal operations. This documentation fell into two categories: (1) actual shipping manifests or electronic databases with information about SDA disposals and (2) documents with process information connected with actual disposals.^{bb}

Data collection for the RPDT relied on several sources to evaluate disposal inventories for fission products, TRU, and activation-product contaminants. The analysis of actinide waste, TRU waste, and fission-product inventories was based on electronic data from RWMIS, hard copies of ANL-W records of remote-handled waste, and hard copies of Form 110 or 135^{cc}. The shipping forms for ANL-W remote-handled waste must be differentiated from standard INEEL Forms 110 and 135. The internal remote-handled waste forms were supplied to INEEL staff by ANL-W. Standardized Form 110s were generated by site-wide INEEL disposal operations. Disposals to SDA pits and soil vaults were recorded in RWMIS. Hard copies of ANL-W remote-handled waste records chronicled soil vault disposals of irradiated subassembly hardware. dd Most of the subassemblies were irradiated. The ANL-W forms covered the years 1984 through 1993.

bb. Related documents are listed in the references section of this report (Section 5).

cc. Standardized U.S. Atomic Energy Form 110/135, "Idaho Operations Office Waste Disposal Request and Authorization" (AEC 1964).

dd. These records also documented shipments to the ANL-W RSWF and the Intermediate-Level Transuranic Storage Facility.

 $\frac{3}{3}$

Table 11. Original and modified descriptions for ANL-W waste streams buried in the SDA from 1984 to 1993.

Original Waste Stream Descriptions ^a			Modified Waste Stream Descriptions	
Waste Stream Number	Description of W	aste	Waste Stream Number	Description of Waste
ANL-704-1R	Contact-handled LLW generated during the fuels in the FMF and Fuel Assembly Storage		ANL-MOD-1R	Soil vault disposals of irradiated subassembly hardware for the years 1984–1993. This stream was characterized by significant amounts activation products
ANL-752-1R	Contact-handled LLW generated during	L&O facility operations.	ANL-MOD-2R	Bulk actinide waste shipments from the ZPPR and other unknown facilities. The principal contaminants consisted of uranium isotopes.
ANL-765-1R	Contact-handled LLW generated during	FCF operations.	ANL-MOD-3R	Fission and actinide waste (some of it fuel bearing) sent to soil vaults for the years 1984–1993.
ANL-765-2R	Remote-handled subassembly LLW genera	ted during FCF operations.	ANL-MOD-4R	General plant waste streams for the years 1984–1993 disposed of in pits. These LLW streams contained small amounts of fission products, actinides, and activation products.
ANL-785-1R	Remote-handled subassembly LLW gene and materials experiments in the HFEF.	rated during nuclear fuel		
ANL-785-2R	Contact-handled LLW generated during	HFEF operations.		
ANL-793-1R	Contact-handled LLW generated during and ZPPR operations.	SCMS, EBR-II, TREAT,		
ANL-798-1R	Contact-handled LLW generated during evaporation of low-level liquid waste in			
	e National Laboratory imental Breeder Reactor-I	HFEF = Hot Fuel Examin L&O = Laboratory and O		SHADE = shielded hot-air drum evaporators TREAT = Transient Reactor Test Facility
	nditioning Facility anufacturing Facility	LLW = low-level waste SCMS = Sodium Compon	nents Maintenance	ZPPR = Zero Power Physics Reactor Shop
a. Original wast	e streams defined as part of the Historical I	Data Task (LMITCO 1995a).		

4.4 Overview of the Analysis Methodology

Table 12 summarizes the analysis methodologies used to analyze each waste stream for the RPDT. The location of the best-estimate inventories for each waste stream is also identified in this table. Additional details about the processes that were used to determine the inventories in each waste stream are discussed in later sections and in the appendices.

Table 12. Summary of the analysis methods used to analyze each ANL-W waste stream for the RPDT.

ANL-W Waste-Streams	Primary Waste Types	Analysis Methods	Location of Best-Estimate Results
ANL-MOD-1R	Irradiated SA hardware	~1,800 SA were identified as being disposed at the SDA. The estimated inventories of activation products were associated with each SA based on its EBR-II core position and benchmark inventories. See Appendix C for analysis details.	Table B-19 and Tables C-2, C-3
ANL-MOD-3R	Fuel-bearing wastes including irradiated and unirradiated dissolved fuel and fuel contaminated materials	Scaling factors were used, along with the estimated mass of heavy metals (from Appendix A) that were disposed at the SDA to estimate the inventory of fission products and actinides that should be presented in these wastes.	Table B-15
ANL-MOD-2R	Low or unirradiated bulk-actinide wastes	The principal contaminants consisted of uranium isotopes and a few other actinides like Pu-239 and Np-237. Inventories were assessed based on the reported shipping data. Except for an estimate of the U-234 inventory (based on the quantity of U-235), no other calculations were made to determine fission product or actinide inventories due to the very low irradiations that these wastes experienced.	Table B-4
ANL-MOD-4R	General plant wastes consisting of small amount of fission products, actinides and activation products	Scaling factors were used along with the reported Cs-137 activities to determine the inventory of fission products, actinides, and activation products that are probably present in these waste items.	Table B-27
SA = subassembly HM = heavy metal			

4.5 Activation Product Inventory

Irradiated subassembly hardware were shipped from EBR-II to the SDA soil vaults from 1977 through 1993. Estimates of activation products recorded in the RWMIS database did not include all required nuclides needed for a risk analysis (e.g., Tc-99 or Ni-59). Consequently, supplemental calculations were made to fill the known gaps in the inventory of activation-products in the RPDT and RWMIS databases. Typical burnup conditions for EBR-II driver fuel rods were employed in a series of reactor physics simulations to calculate the buildup of activation products. These simulations generated approximate activation-product activity levels for subassembly components sent to the RWMC.

The EBR-II core comprised a hexagonal lattice of various subassembly types. Several generations of fuel designs (e.g., Mark-I, Mark-IA, Mark-II, and Mark-III) were used in EBR-II, and the burnups varied considerably over individual recorded histories. Subcategories of these different components included such subassembly classifications as core or driver fuels and inner and outer blanket. These defueled subassembly components usually included top and bottom reflector pieces and a cylindrical, hexagonal outer shell that surrounded the fuel pins. In addition, irradiated control rod assemblies also were included in the waste streams sent to the RWMC. The materials composition of the subassembly components was either 304 or 316 stainless steel.

Records from the RWMIS show hundreds of waste shipments with small amounts of activation-product contaminants were sent to the SDA pits. However, the net activated-product pit disposals were less than 3% of the activity contained within the subassembly hardware from EBR-II. As in later shipments made during the RPDT period, the subassembly components consisted of highly irradiated 304 and 316 stainless steel alloys. The best-estimate RPDT inventories of activation products are shown in Table 13 and compared with information from the RWMIS and RPDT. Finally, an overall comparison is shown in Table 14 for the combined period of 1952 through 1993. Note that the corresponding HDT data are shown in Table 4.

Table 13. Comparison of activation-product activities for the years 1984 through 1993.

Nuclide	RWMIS Reported Total Activity (Ci)	RPDT Reported Total Activity (Ci)	Best-Estimate Activity ^a (Ci)
C-14	_	-	15.4
C1-36	_	_	0.0038
Co-60	717,000	1,100,000	735,000
Ni-59	_	_	78.9
Ni-63	_	_	5,730
Nb-94	_	_	2.55
Tc-99	_	_	6.24 ^b

RPDT = Recent and Projected Data Task (LMITCO 1995b)

RWMIS = Radioactive Waste Management Information System

b. This value represents the Tc-99 inventory in subassembly hardware only. A contribution also exists in fission product contaminated wastes.

-

a. Obtained from Table B-18.

ee. After 1993, irradiated hardware was stored in concrete-lined silos at the RWMC.

Table 14. Overall comparison of activation-product activities for the years 1952 through 1993.

	RWMIS Reported Total Activity	RPDT Reported Total Activity	Best-Estimate Activity ^a
Nuclide	(Ci)	(Ci)	(Ci)
C-14	_	_	3.15E+01
Cl-36	_	_	7.90E-03
Co-60	1.23E+05	1.43E+06	1.54E+06
Ni-59	_	_	1.65E+02
Ni-63	_	_	1.20E+04
Nb-94	_	_	5.38E+00
Tc-99	<u> </u>	_	$13.1 \text{ E} + 00^{\text{b}}$

RPDT = Recent and Projected Data Task (LMITCO 1995b)

RWMIS = Radioactive Waste Management Information System

4.6 Estimated Actinide Inventory

Disposals of both irradiated and unirradiated actinide (e.g., heavy metal) wastes were identified in shipping records from 1960 through 1988. As previously mentioned, no data were found regarding ANL-W waste shipments prior to 1960. The radioactive waste streams that were identified included dissolved (irradiated and unirradiated) fuel samples and bulk-actinide disposals of both depleted and natural uranium. In general, these waste streams fall into two categories: (1) sporadic shipments generally having significant bulk weights of unirradiated or low-irradiated actinide waste (mainly uranium); and (2) highly irradiated fuel samples or irradiated test specimens with weights that were generally less than 100 g (not including the container weight). However, some shipments contained unirradiated fuel specimens as well.

The best-estimate activity data corresponding to the reported (unirradiated) uranium mass data shown in Table B-4 are listed in Table 15. The total heavy metal mass that was disposed at the SDA for irradiated fuel-bearing wastes are shown in Table 16.

Because the irradiation histories associated with bulk-actinide wastes (in Table 15) were very low, fission product and TRU isotopes resulting from fission or capture events were considered negligible for waste streams ANL-MOD-4H (1952 through 1983) and ANL-MOD-2R (1984 through 1993). Therefore, for the bulk-actinide wastes, no fission products or TRU isotopes (beyond those isotopes already reported by the waste generator) were estimated to exist with these waste streams (see Tables B-1 and B-4). However, in the case of fuel-bearing waste shipments, as illustrated in Table 15 (obtained from Appendix A), these wastes usually consisted of irradiated fuel specimens or similar material (e.g., irradiated flux wires, etc.). In this case, calculations were made (based on scaling factors) to predict the inventory of fission products and TRU that probably were present in these heavy-metal wastes, but were generally not reported. These calculations were applied to waste streams ANL-MOD-3H and ANL-MOD-2H, but not to waste stream ANL-MOD-2R (and ANL-MOD-4H). These calculations are discussed in greater detail in Section B-3 of Appendix B.

a. Obtained from Table B-18 in Appendix B.

b. This value represents the Tc-99 inventory in subassembly hardware only. A contribution of 3.4 Ci also exists in fission product wastes. The total Tc-99 inventory, taken from Table B-31 (Part 5) is 16.5 Ci.

Table 15. Best-estimate mass inventory of irradiated uranium wastes sent to the SDA from 1984 through 1993 (e.g., during the RPDT) and are included in the ANL-MOD-2R waste-stream.

SDA Disposal Year	U-234 ^a (kg)	U-235 (kg)	U-238 (kg)	Ratio of U234 to U235	Total Uranium (kg)
1984	6.85E-03	8.97E-01	1.04E+02	7.64E-03	1.05E+02
1985	1.49E-03	1.96E-01	3.26E-01	7.64E-03	5.23E-01
1986	1.04E-02	1.37E+00	1.24E+01	7.64E-03	1.38E+01
1987	4.46E-02	5.84E+00	9.52E+01	7.64E-03	1.01E+02
1988	6.23E-02	8.16E+00	1.79E+01	7.64E-03	2.62E+01
1989	1.84E-02	2.41E+00	9.53E+00	7.64E-03	1.20E+01
1990	2.52E-03	3.31E-01	1.58E-01	7.64E-03	4.91E-01
1991	9.27E-04	1.21E-01	3.48E-01	7.64E-03	4.70E-01
1992	2.95E-03	3.86E-01	1.13E+01	7.64E-03	1.17E+01
1993	4.92E-03	6.44E-01	2.19E-01	7.64E-03	8.68E-01
Total	1.55E-01	2.03E+01	2.51E+02		2.72E+02

a. The U-234 inventory was not reported. Instead, the U-234 inventory was determined based on the U-235 mass and the natural ratio of U-234 to U-235, which is 7.64E-03.

Table 16. Shipping data for irradiated fuel-bearing wastes (e.g., fuel samples) sent to the SDA from 1984 through 1993.^a

D' 1	Net Actinide or Heavy Metal Mass Disposed at the SDA ^b	Reported Activity that was Shipped ^c
Disposal	*	
Year	(kg)	(Ci)
1984	-	_
1985	2.79E-02	4.9E+03
1986	9.14E-02	1.9E+02
1987	4.27E-01	1.4E+04
1988	1.58E+01	5.5E+03
1989	_	
1990	_	
1991	_	_
1992	_	
1993	_	
Total	1.63E+01	2.5E+04

a. Note: no data exists for the period 1952 through 1959. See Table 6 for the corresponding information for the HDT.

b. Obtained from the underlined "total" values shown in Tables A-1 through A-25 of Appendix A.

c. The activity data were reported on the Form 110-shipping manifests and may not correlate with curies calculated from the individual radionuclides. See Appendix A for a detailed list of radionuclides.

The actinide data corresponding to the detailed analysis presented in Appendix B are summarized in Tables 17 and 18. These data are also compared with similar data from the RPDT (LMITCO 1995b). Note that the inventories of uranium isotopes are in reasonable agreement (within a factor of 3). However, the amounts of Th-232 and Pu-239 are higher than the corresponding RPDT values. Differences in the Pu-239 activities appear to originate from the inclusion of new data based on Form 110 shipping records (i.e., three shipments of low-irradiated actinide waste). Finally, there are no data for Np-237 in the HDT (LMITCO 1995a) or RPDT (LMITCO 1995b) to compare against.

Table 17. Best-estimate actinide disposals made at the SDA from 1984 through 1993.

	Best-estimate Inventory ^a for Bulk Actinides	RPDT Inventory for Bulk Actinides
Nuclide	(Ci)	(Ci)
Th-232	1.13E-04	_
U-234	1.23E+00	2.5E+00
U-235	5.47E-02	1.1E-01
U-238	8.65E-02	5.3E-01
Pu-239	7.24E+01	1.6E+00
Np-237	3.51E-03	_

RPDT = Recent and Projected Data Task (LMITCO 1995b) a. Obtained from Table B-31 (Part 5).

Table 18. Total actinide disposals made at the SDA from 1960 through 1993.^a

Nuclide	Total Inventory ^b for Bulk-actinides (Ci)	RPDT Total Inventory for Bulk-actinides (Ci)
Th-232	9.62E-04	1.0E-05
U-234	3.39E+00	5.9E+00
U-235	1.47E-01	3.8E-01
U-238	1.39E+00	1.73E+00
Pu-239	$5.11E+02^{c}$	1.26E+01
Np-237	2.67E-02	_

RPDT = Recent and Projected Data Task (LMITCO 1995b)

Similar to the calculation of fission products present in general plant wastes, due to the presence of Cs-137; actinides are expected to exist in waste streams that contain Cs-137 (or those waste streams that contain irradiated heavy metals). In other words, Cs-137 is probably present because it is associated with irradiated fuel particles; and therefore, fission products, uranium, and actinides should also be present. Details regarding the estimated inventory of actinides present in waste streams associated with Cs-137 can be found in Appendix B (see Tables B-9, B-12, B-15, and B-24).

a. No data exists for the period 1952–1959. Since disposals from ANL-W to the SDA would have been small during this period, then the above results can also be applied to 1952 through 1993.

b. Obtained from Table B-31 (Part 5).

c. Some records appear to indicate that a shipment of 2.4 kg (149 Ci) of plutonium with 25 EBR-I fuel rods was disposed at the SDA in 1964.

The presence of unirradiated actinide contaminants is separately factored into the total inventory. In addition, this method may not be directly applicable to contaminants from specialized irradiation experiments that have irradiation histories and compositions that deviate significantly from standard EBR-II driver fuel characteristics.

Explicitly identified waste streams with actinide contaminants (amounts reported by weight) from remote shipping records are thought to be irradiated fuel samples. Most SDA hardware shipments listing a Pu-239/240 value are calculated from hot cell smears. Waste descriptions for RWMIS pit shipments for 1984–1993 were encoded with generic identification numbers. The RWMIS database did not always clearly identify the process that generated these actinide waste streams or associated disposed material states (e.g., depleted uranium, natural uranium, and oxide).

4.7 Estimated Fission Product Inventory

Fission product inventories were estimated to be present in the following RPDT fuel-bearing waste stream: ANL-MOD-3R, and in the general plant waste stream: ANL-MOD-4R. As previously mentioned, fission products were estimated to be present in fuel-bearing wastes because of the heavy-metal mass inventories (usually uranium) and the fact that these wastes generally originated from irradiated fuel. Fission products were estimated to be present in general plant wastes due to the presence of Cs-137. The estimated inventories of fission products in these two categories was accomplished in similar ways. For the fuel-bearing wastes, the inventory of unreported fission products (or activation products) was estimated by multiplying scaling factors (Ci/kg-HM) by the reported heavy metal inventory (kg). For general plant wastes, the inventory of unreported radionuclides was estimated by multiplying scaling factors based on Cs-137 (Ci/Ci-Cs137) by the estimated Cs-137 activity (Ci) in this waste.

Estimates of many fission products (except Cs-137) were calculated using scaling factor data. That is, an average isotopic profile was used to quantify the activities for the RPDT period. A single gamma emitter was selected from which all other radionuclides could be determined. In this report, Cs-137 was chosen as the reference isotope because it can be easily detected and has been consistently reported in many RWMIS shipments. Logged Cs-137 activities constituted a significant fraction of the total reported MFP activities. Net Cs-137 gamma activities were then scaled to calculate the activity of other nuclides of interest—such as I-129—that were not explicitly reported.

The scaling method used in this study is based on highly irradiated EBR-II driver fuel. Representative scaling factors for a typical EBR-II driver fuel element are listed in Appendix B (see Table B-8). Again, the derived scaling factors are referenced to Cs-137, and are ultimately based on reactor physics calculations of the inventory present in EBR-II driver fuel pins at maximum burnup conditions (see Section B-3 of Appendix B for additional details). Waste associated with experimental fuel assemblies would have been generated during destructive examination procedures. The isotopic profile associated with experimental assemblies or other irradiated fuel capsules may differ from the profile from EBR-II driver fuel. Consequently, some uncertainties are introduced by applying one set of scaling factors

39

ff. A significant fraction of the disposed fission products and TRU contaminants were connected with EBR-II operations: processes for handling irradiated fuel and decontamination. These process waste streams would have included disposals of LLW connected with hot cell operation and decontamination. Other minor waste streams containing fission products contaminants originated from TREAT, ZPPR, and other support facilities. Contributions from support facilities were typically not as significant as from EBR-II.

gg. This mode of contaminant reporting was applicable only to waste streams contaminated with fission products. In addition, isotopic breakdowns of reported fission product contaminants were not always broken out. In many older shipping manifests, only net activities for the total amount of MFP were reported.

hh. Examples of destructive examination include chemical dissolution of irradiated fuel or dissection of fuel in hot cells.

to all waste shipments. However, since the vast majority of all fuel that processed (or reprocessed) at ANL-W facilities was from EBR-II driver fuel assemblies, this assumption is probably reasonable.

Waste streams containing fission products (and actinides) were buried in both pits and soil vaults. A detailed yearly breakdown of activities disposed at the SDA is shown in Appendix B. Activity totals are summarized in Table 19 for selected fission products and were obtained from Table B-31 (Part 5). This data is also compared against information for the RPDT (LMITCO 1995b).

Table 19. Summary of the total fission product waste activities that were disposed at the SDA from ANL-W operations for 1984 through 1993 (obtained from Table B-31).

Nuclide	Best-estimate Activity (Ci)	RPDT Activity (Ci)
H-3	2.14E+01	8.2E+01
Sr-90	3.16E+03	1.6E+02
Tc-99	$6.77E+00^{a}$	<u> </u>
I-129	1.10E-03	_
Cs-137	4.37E+03	1.5E+02
Eu-152	2.22E-01	_
Eu-154	2.32E+01	5.8E-01
Total	7.58E+03	3.9E+02

a. Tc-99 exists as both a fission product and activation product. The listed number combines both components. The amount of Tc-99 from activated metal (in ANL-MOD-1R) is 0.64 Ci (see Table B-19).

4.8 Uncertainties

In general, the uncertainty information presented in Section 3.8 for the HDT period also applies to the RPDT data. Nevertheless, some of this information is repeated below.

Because of the highly variable and shipment-dependent nature of many of the waste streams, standard statistical uncertainty methods were not feasible to define individual uncertainty factors. As in the original HDT and RPDT reports (LMITCO 1995a, 1995b), the methodology for defining the best-estimate activities and associated upper and lower bounds was not based on a rigorous statistical error propagation model. Instead, approximation methods were based on professional (e.g., engineering) judgment and other reasonable assumptions.

Based on engineering judgment, an uncertainty factor of <u>at least</u> 1.5 should be applied to the estimated activity of all radionuclides. However, in some cases, uncertainty factors might be as high as a factor of 100. In the case of the bulk-actinide waste stream: ANL-MOD-2R, an uncertainty factor of 1.50 was assumed for all radionuclides. That is, for this waste stream, the Upper-bound activities were estimated by multiplying the best-estimate activities by a factor of 1.50; and the Lower-bound activities were estimated by dividing the best-estimate activities by 1.50. See Tables B-1 through B-5 for additional details.

In the case of waste streams activities that were estimated with the aid of scaling factors, then individual uncertainty factors were applied to each radionuclide. These factors ranged from 1.1 for U-235 up to a factor of 100 for Cl-36. Additional details are shown in Table B-7 of Appendix B. See Section 3.8 for additional details concerning the uncertainty analysis.

5. CONCLUSIONS AND RECOMMENDATIONS

This report documents the reassessment inventory of ANL-W waste disposal shipments sent to the SDA from 1952 through 1993. Reassessments of waste streams consisting of fission products, activation products, and actinide waste are reported. Generally, documentation for shipments after 1983 was more complete than earlier shipments. The total activity estimates for activation products, fission products, and actinide contaminants for the HDT and RPDT periods (1960" –1993) are summarized in Table 20. Detailed inventory data are shown in Appendix B. In addition to best-estimate data, Table 20 also incorporates lower-bound and upper-bound activities. The information in Table 20 indicates that ~96% of the total activity of all radionuclides disposed at the SDA from ANL-W is due to Co60, and this comes from activated subassembly steel. The next most important contributor to the total activity is Cs-137.

The data for waste stream ANL-MOD-2Hext is simply the ANL-MOD-2H waste stream extended into the RPDT period as a convenience because it was contained in the HDT data set. (See Table B-12).

Definitions of lower-bound, best-estimate, and upper-bound inventories were not developed with standard statistical methods. The methodology for defining the upper-bound and lower-bound activities was based on professional judgment and other reasonable assumptions. The rationale for not using rigorous statistical methods was due to the highly variable nature of the waste disposals and the information found in the disposal records that made such methods unfeasible.

The best-estimate analysis of activation-product disposals associated with irradiated subassembly hardware was judged as acceptable for two reasons. First, documentation of soil vault disposals of activated products showed that all of the associated shipping records and supporting documentation had been located. Personnel from ANL-W, working in conjunction with INEEL staff, located these shipping records. Second, extensive deterministic calculations generated technically defensible estimates of contaminant disposals for each disposed subassembly. The details concerning the subassembly hardware analysis are documented in Appendix C. In contrast, estimated inventories of fission products and actinide waste contaminants for major waste streams have larger uncertainties. Reduction in these uncertainties may be possible if additional information becomes available.

Tables 21 and 22 show the activity and relative distributions of radionuclides among the possible waste streams. Based on these data, it is clear that ~97% of the total disposed activity is associated with activated subassembly hardware found in the two waste streams: ANL-MOD-1H and ANL-MOD-1R. Most of this activity results from Co-60. Finally, it can be seen that the majority of fission product and actinide activities are contained in the General Plant wastes found in waste stream ANL-MOD-5H.

_

ii. This period also represents 1952 through 1993 since there are no data from 1952 through 1959.

Table 20. Summary of the total inventory of activation products, fission products, and actinide waste activities that were disposed at the SDA from ANL-W for 1960 through 1993.^a

Nuclide	Lower Bound (Ci)	Best-estimate (Ci)	Upper Bound (Ci)						
H-3	3.61E+01	1.49E+02	7.01E+02						
C-14	1.57E+01	3.15E+01	6.33E+01						
C1-36	3.94E-03	7.98E-03	2.72E-02						
Co-60	7.72E+05	1.54E+06	3.09E+06						
Ni-59	8.23E+01	1.65E+02	3.32E+02						
Ni-63	6.01E+03	1.20E+04	2.40E+04						
Sr-90	1.00E+04	2.01E+04	4.01E+04						
Nb-94	2.69E+00	5.38E+00	1.08E+01						
Tc-99	8.26E+00	1.65E+01	3.30E+01						
I-129	3.50E-03	7.01E-03	1.40E-02						
Cs-137	1.39E+04	2.78E+04	5.55E+04						
Eu-152	2.82E-01	1.41E+00	7.05E+00						
Eu-154	2.94E+01	1.47E+02	7.36E+02						
Ra-226	1.33E-01	2.00E-01	3.00E-01						
Th-232	3.05E-04	9.62E-04	3.96E-03						
U-234	1.98E+00	3.39E+00	5.93E+00						
U-235	1.14E-01	1.47E-01	1.93E-01						
U-236	5.40E-02	1.08E-01	2.16E-01						
U-238	9.23E-01	1.39E+00	2.09E+00						
Np-237	1.41E-02	2.67E-02	5.12E-02						
Pu-238	3.84E+00	1.15E+01	3.46E+01						
Pu-239	2.64E+02	5.11E+02	9.97E+02						
Pu-240	7.04E-01	7.04E+00	7.04E+01						
Pu-241	2.42E+00	1.21E+02	6.06E+03						
Pu-242	1.94E-04	1.94E-03	1.94E-02						
Am-241	3.03E-01	3.03E+00	3.03E+01						
Total	8.02E+05	1.60E+06	3.22E+06						
a. Obtained from Table	Obtained from Table B-34 of Appendix B.								

Table 21. Best-estimate activity distribution of radionuclides within the ten possible waste streams based on the total curie inventory (1960 through 1993).

											-
Nuclide	MOD-1H	MOD-2H	MOD-2Hext	MOD-3H	MOD-4H	MOD-5H	MOD-1R	MOD-2R	MOD-3R	MOD-4R	Total
H-3	0.00E+00	2.36E+01	1.97E+01	2.09E+01	1.33E+01	6.83E+01	0.00E+00	0.00E+00	3.22E+00	4.95E-01	1.49E+02
C-14	1.60E+01	1.61E-02	1.34E-02	1.42E-02	0.00E+00	4.65E-02	1.54E+01	0.00E+00	2.19E-03	3.37E-04	3.15E+01
CL-36	4.05E-03	1.99E-05	1.65E-05	1.76E-05	0.00E+00	5.74E-05	3.82E-03	0.00E+00	2.70E-06	4.16E-07	7.98E-03
CO-60	8.09E+05	9.23E+01	7.68E+01	8.16E+01	0.00E+00	2.67E+02	7.35E+05	0.00E+00	1.26E+01	1.93E+00	1.54E+06
NI-59	8.57E+01	4.83E-02	4.02E-02	4.27E-02	0.00E+00	1.39E-01	7.89E+01	0.00E+00	6.57E-03	1.01E-03	1.65E+02
NI-63	6.27E+03	1.86E+00	1.55E+00	1.65E+00	0.00E+00	5.38E+00	5.73E+03	0.00E+00	2.53E-01	3.90E-02	1.20E+04
SR-90	0.00E+00	3.48E+03	2.90E+03	3.08E+03	0.00E+00	1.01E+04	0.00E+00	0.00E+00	4.74E+02	7.30E+01	2.01E+04
NB-94	2.81E+00	2.66E-03	2.21E-03	2.35E-03	0.00E+00	7.68E-03	2.55E+00	0.00E+00	3.61E-04	5.57E-05	5.38E+00
TC-99	6.88E+00	5.89E-01	4.90E-01	5.21E-01	0.00E+00	1.70E+00	6.24E+00	0.00E+00	8.01E-02	1.23E-02	1.65E+01
I-129	0.00E+00	1.22E-03	1.01E-03	1.07E-03	0.00E+00	3.51E-03	0.00E+00	0.00E+00	1.65E-04	2.55E-05	7.01E-03
CS-137	0.00E+00	4.82E+03	4.01E+03	4.26E+03	0.00E+00	1.39E+04	0.00E+00	0.00E+00	6.55E+02	1.01E+02	2.78E+04
EU-152	0.00E+00	2.45E-01	2.04E-01	2.16E-01	0.00E+00	7.07E-01	0.00E+00	0.00E+00	3.33E-02	5.13E-03	1.41E+00
EU-154	0.00E+00	2.55E+01	2.12E+01	2.26E+01	0.00E+00	7.38E+01	0.00E+00	0.00E+00	3.47E+00	5.35E-01	1.47E+02
RA-226	0.00E+00	5.18E-08	4.31E-08	4.58E-08	2.00E-01	1.50E-07	0.00E+00	0.00E+00	7.04E-09	1.08E-09	2.00E-01
TH-232	0.00E+00	1.25E-04	1.04E-04	1.11E-04	2.42E-04	3.61E-04	0.00E+00	0.00E+00	1.70E-05	2.62E-06	9.62E-04
U-234	0.00E+00	2.91E-01	2.42E-01	2.57E-01	7.48E-01	8.40E-01	0.00E+00	9.71E-01	3.96E-02	6.09E-03	3.39E+00
U-235	0.00E+00	1.19E-02	9.87E-03	1.05E-02	3.42E-02	3.43E-02	0.00E+00	4.40E-02	1.61E-03	2.49E-04	1.47E-01
U-236	0.00E+00	1.87E-02	1.56E-02	1.66E-02	0.00E+00	5.41E-02	0.00E+00	0.00E+00	2.55E-03	3.92E-04	1.08E-01
U-238	0.00E+00	2.28E-03	1.90E-03	2.01E-03	1.29E+00	6.58E-03	0.00E+00	8.44E-02	3.10E-04	4.77E-05	1.39E+00
NP-237	0.00E+00	3.88E-03	3.23E-03	3.43E-03	4.37E-03	1.12E-02	0.00E+00	0.00E+00	5.27E-04	8.12E-05	2.67E-02
PU-238	0.00E+00	2.00E+00	1.66E+00	1.77E+00	0.00E+00	5.78E+00	0.00E+00	0.00E+00	2.72E-01	4.19E-02	1.15E+01
PU-239	0.00E+00	7.98E+01	6.64E+01	7.06E+01	5.15E+01	2.31E+02	0.00E+00	0.00E+00	1.09E+01	1.67E+00	5.11E+02
PU-240	0.00E+00	1.22E+00	1.02E+00	1.08E+00	0.00E+00	3.53E+00	0.00E+00	0.00E+00	1.66E-01	2.56E-02	7.04E+00
PU-241	0.00E+00	2.10E+01	1.75E+01	1.86E+01	0.00E+00	6.07E+01	0.00E+00	0.00E+00	2.86E+00	4.40E-01	1.21E+02
PU-242	0.00E+00	3.36E-04	2.80E-04	2.97E-04	0.00E+00	9.71E-04	0.00E+00	0.00E+00	4.57E-05	7.04E-06	1.94E-03
Pu-244	0.00E+00	2.22E-13	1.85E-13	1.96E-13	0.00E+00	6.42E-13	0.00E+00	0.00E+00	3.02E-14	4.66E-15	1.28E-12
AM-241	0.00E+00	5.27E-01	4.38E-01	4.65E-01	0.00E+00	1.52E+00	0.00E+00	0.00E+00	7.16E-02	1.10E-02	3.03E+00
Totals =	8.15E+05	8.55E+03	7.11E+03	7.56E+03	6.71E+01	2.47E+04	7.41E+05	1.10E+00	1.16E+03	1.79E+02	1.60E+06

4

Table 22. Best-estimate percent distribution of curie inventory within the ten possible waste streams (1960 through 1993).

		1			J						
Nuclide	MOD-1H	MOD-2H	MOD-2Hext	MOD-3H	MOD-4H	MOD-5H	MOD-1R	MOD-2R	MOD-3R	MOD-4R	Total
H-3	0.00%	15.81%	13.16%	13.97%	8.90%	45.68%	0.00%	0.00%	2.15%	0.33%	100.00%
C-14	50.95%	0.05%	0.04%	0.05%	0.00%	0.15%	48.76%	0.00%	0.01%	0.00%	100.00%
CL-36	50.78%	0.25%	0.21%	0.22%	0.00%	0.72%	47.78%	0.00%	0.03%	0.01%	100.00%
CO-60	52.37%	0.01%	0.00%	0.01%	0.00%	0.02%	47.59%	0.00%	0.00%	0.00%	100.00%
NI-59	51.98%	0.03%	0.02%	0.03%	0.00%	0.08%	47.85%	0.00%	0.00%	0.00%	100.00%
NI-63	52.22%	0.02%	0.01%	0.01%	0.00%	0.04%	47.69%	0.00%	0.00%	0.00%	100.00%
SR-90	0.00%	17.36%	14.44%	15.34%	0.00%	50.14%	0.00%	0.00%	2.36%	0.36%	100.00%
NB-94	52.26%	0.05%	0.04%	0.04%	0.00%	0.14%	47.45%	0.00%	0.01%	0.00%	100.00%
TC-99	41.68%	3.57%	2.97%	3.15%	0.00%	10.30%	37.77%	0.00%	0.49%	0.07%	100.00%
I-129	0.00%	17.36%	14.44%	15.34%	0.00%	50.14%	0.00%	0.00%	2.36%	0.36%	100.00%
CS-137	0.00%	17.36%	14.44%	15.34%	0.00%	50.14%	0.00%	0.00%	2.36%	0.36%	100.00%
EU-152	0.00%	17.36%	14.44%	15.34%	0.00%	50.14%	0.00%	0.00%	2.36%	0.36%	100.00%
EU-154	0.00%	17.36%	14.44%	15.34%	0.00%	50.14%	0.00%	0.00%	2.36%	0.36%	100.00%
RA-226	0.00%	0.00%	0.00%	0.00%	100.00%	0.00%	0.00%	0.00%	0.00%	0.00%	100.00%
TH-232	0.00%	12.99%	10.81%	11.48%	25.15%	37.52%	0.00%	0.00%	1.77%	0.27%	100.00%
U-234	0.00%	8.57%	7.13%	7.57%	22.04%	24.75%	0.00%	28.60%	1.17%	0.18%	100.00%
U-235	0.00%	8.10%	6.73%	7.15%	23.36%	23.39%	0.00%	30.00%	1.10%	0.17%	100.00%
U-236	0.00%	17.36%	14.44%	15.34%	0.00%	50.14%	0.00%	0.00%	2.36%	0.36%	100.00%
U-238	0.00%	0.16%	0.14%	0.15%	92.97%	0.47%	0.00%	6.08%	0.02%	0.00%	100.00%
NP-237	0.00%	14.52%	12.08%	12.83%	16.36%	41.93%	0.00%	0.00%	1.97%	0.30%	100.00%
PU-238	0.00%	17.36%	14.44%	15.34%	0.00%	50.14%	0.00%	0.00%	2.36%	0.36%	100.00%
PU-239	0.00%	15.61%	12.99%	13.79%	10.08%	45.08%	0.00%	0.00%	2.12%	0.33%	100.00%
PU-240	0.00%	17.36%	14.44%	15.34%	0.00%	50.14%	0.00%	0.00%	2.36%	0.36%	100.00%
PU-241	0.00%	17.36%	14.44%	15.34%	0.00%	50.14%	0.00%	0.00%	2.36%	0.36%	100.00%
PU-242	0.00%	17.36%	14.44%	15.34%	0.00%	50.14%	0.00%	0.00%	2.36%	0.36%	100.00%
AM-241	0.00%	17.36%	14.44%	15.34%	0.00%	50.14%	0.00%	0.00%	2.36%	0.36%	100.00%
All isotopes	50.78%	0.53%	0.44%	0.47%	0.00%	1.54%	46.15%	0.00%	0.07%	0.01%	100.00%

6. REFERENCES

- 42 USC § 9601 et seq., 1980, "Comprehensive Environmental Response, Compensation and Liability Act of 1980 (CERCLA/Superfund)," *United States Code*.
- AEC Form 110, 1964, "Waste Disposal Request and Authorization," U.S. Atomic Energy Commission Idaho Operations Office, Idaho Falls, Idaho, November 1964.
- Benedict, M., T. H. Pigford, and H. W. Levi, 1981, *Nuclear Chemical Engineering*, New York: McGraw-Hill College Division.
- Brown, B. W., D. L. Crandall, R. E. Dafoe, M. R. Dolenc, and D. M. LaRue, 1987, *Functional and Operational Design Requirements for Decontamination and Decommissioning of the EBR-I Mark II NaK*, EGG-WM-7264, Rev. 0, Idaho National Engineering and Environmental Laboratory.
- Case, M. J., A. S. Rood, J. M. McCarthy, S. O. Magnuson, B. H. Becker, and T. K. Honeycutt, 2000, Technical Revision of the Radioactive Waste Management Complex Low-Level Waste Radiological Performance Assessment for Calendar Year 2000, INEEL/EXT-00-01089, Idaho National Engineering and Environmental Laboratory.
- EG&G, 1985, A History of the Radioactive Waste Management Complex at the Idaho National Engineering Laboratory, WM-F1-81-003, Rev. 3, Idaho National Engineering and Environmental Laboratory, July 1985.
- Haroldsen, R. O., F. D. McGinis, M. Novick, R. R. Smith, and F. W. Thalgott, 1961, *Hazards Summary Report EBR-I Mark IV*, ANL-6411, Argonne National Laboratory-West.
- Holdren, K. Jean and Barbara J. Broomfield, 2003, Second Revision to the Scope of Work for the Operable Unit 7-13/14 Waste Area Group 7 Comprehensive Remedial Investigation/Feasibility Study, INEL-95/0253, Rev. 2, Idaho National Engineering and Environmental Laboratory.
- Holdren, K. Jean, Bruce H. Becker, Nancy L. Hampton, L. Don Koeppen, Swen O. Magnuson, T. J. Meyer, Gail L. Olson, and A. Jeffrey Sondrup, 2002, *Ancillary Basis for Risk Analysis of the Subsurface Disposal Area*, INEEL/EXT-02-01125, Rev. 0, Idaho National Engineering and Environmental Laboratory, September 2002.
- INEEL, 2002, "Environmental Restoration Optical Imaging System Retrieval," URL: http://zeus.inel.gov:8080/docs/ois/ois_index.html
- Kendall, E. W. and D. K. Wang, 1975, *Decontamination and Decommissioning of the EBR-I Complex Final Report*, ANCR-1242, Rev. 0, Idaho National Engineering and Environmental Laboratory.
- Kittel, J. H., M. Novick, and R. F. Buchanan, 1957, *The EBR-I Meltdown—Physical and Metallurgical Changes in the Core*, ANL-5731, Rev. 0, Argonne National Laboratory-West.
- Koch, L. J., W. B. Loewenstein, and H. O. Monson, 1962, *Addendum to Hazard Summary Report Experimental Breeder Reactor-II (EBR-II)*, ANL-5719, Argonne National Laboratory-West.

- Little, M., J. K. McDonald, J. D. Grandy, M. L. Carboneau, A. G. Chambers, S. C. Flynn, L. D. Koeppen, and J. A. Logan, 2001, *A Comprehensive Inventory of Radiological and Nonradiological Contaminants in Waste Buried or Projected to Be Buried in the Subsurface Disposal Area of the INEEL RWMC During the Years 1984 to 2003 Supplement*, INEL-95/0135 Supplement, Rev. 0, Idaho National Engineering and Environmental Laboratory.
- LMITCO, 1995a, A Comprehensive Inventory of Radiological and Nonradiological Contaminants in Waste Buried in the Subsurface Disposal Area of the INEL RWMC During the Years 1952-1983, INEL-95/0310, Rev. 1, Idaho National Engineering and Environmental Laboratory.
- LMITCO, 1995b, A Comprehensive Inventory of Radiological and Nonradiological Contaminants in Waste Buried or Projected to Be Buried in the Subsurface Disposal Area of the INEL RWMC During the Years 1984-2003, INEL-95/0135, Rev. 1, Idaho National Engineering and Environmental Laboratory.
- Smith, R. R., R. O. Haroldsen, R. E. Horne, and R. G. Matlock, 1961, *The Breeding Ratio of a Plutonium Loading in EBR-I*, ANL-6789, Argonne National Laboratory-West.
- Stevenson, C. E., 1987, The EBR-II Fuel Cycle Story, LaGrange, Illinois: American Nuclear Society.
- Witbeck, L. C., and R. M. Fryer, 1979, *Waste Production and Management at EBR-II*, ANL-79-14, Argonne National Laboratory-West.